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On the atomic volume relations in certain isomorphous series.

By A. F. Hallimond, M.A., F.G.S.

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[Read June 14, 1927.]

CONSIDERABLE interest attaches to the estimation of the volume occupied by the several atoms in a crystalline compound. For the complete molecule a volume can at once be calculated from the density and the chemical composition, but the further subdivision of this space into domains belonging to the constituent atoms or groups can only be made by methods that are limited and frequently very indirect. In general, the volume is not additive, and it seems clear that the same atom may occupy very different volumes in its different compounds; indeed, it is only within the limits of a truly isomorphous series that the volume can be treated as an additive property. But even in this case it is not possible directly to calculate volumes for the constituents of the molecule if the chemical formulae are of the same type throughout; when, however, the formulae differ in type a solution may be possible. In this way approximate values have been obtained for the volumes of the several component oxides of the mica group,[2] and it seemed desirable

[1] Communicated by permission of the Director.

[2] A. F. Hallimond, Min. Mag., 1927, vol. 21, pp. 195–204.

to examine some of the better known isomorphous series in order to see how far the conclusions to be drawn from the latter would correspond with those based on the poorer data available for the micas. By far the best available series are those containing the alkali metals, for many of which data of the highest accuracy have been supplied in the extensive researches of Tutton and other workers. In these salts the formulae are strictly analogous, so that the direct method which served for the micas is not available ; some other hypothesis must form a basis for calculating the atomic volumes, and the present paper contains a discussion of the data from this standpoint. The several isomorphous salt-series formed by the eutropic metals potassium, rubidium, and caesium are characterized by an approximate constancy in the ratio between the volume-differences produced by interchange of the metals. Further, the volumes of the free metals yield the same ratio, so that the metals themselves apparently constitute an isomorphous series quite analogous with the sulphates, selenates, and other salts. The differences vary in size in the different series ; a direct additive relation does not hold good throughout, but the volume of the combined metal appears to be derived from that of the free metal by a factor common to each salt-series. In this way volumes can be calculated for the metal and acid radicle in each salt. A similar constant ratio holds good for the chlorides, bromides, and iodides ; in the absence of a solid isomorphous series for the free halogens a set of comparable volumes can be obtained from the partial volumes calculated for the alkali halides ; these agree with the values obtained by Richards from the compressibility, and are nearly proportional to those given by Kopp's method of comparing the volume of the liquids at the boiling-point. By the use of these and similar data estimates can be obtained for the volume of the atoms and radicles in numerous compounds of the alkali metals and the halogens, and these estimates agree well with the approximate values generally accepted.

A. *Isomorphous series derived from the eutropic metals K, Rb, Cs.*

Some of the most accurately known data for the alkali salts are collected in Table I. The first three columns contain the molecular volumes of the salts ; the next two columns give the values for the differences produced by the eutropic replacement of K by Rb and by Cs, and the fifth column shows the ratio between these differences. Although the data exhibit a wide variety in molecular volume, in composition, and

in the magnitude of the differences, yet the ratio remains very closely constant throughout, and has the same value even in the case of the free metals, though these have a greater volume than the whole of the sulphate molecule.

Table I. Molecular volumes of isomorphous salts of K, Rb, Cs.

	K.	Rb.	Cs.	Rb–K.	Cs–K.	$\frac{\text{Cs–K}}{\text{Rb–K}}$
Free metal M_2 [1] ...	90·8	111·8	141·0	21·0	50·2	2·39
Sulphate [2]	64·91	73·34	84·58	8·43	19·67	2·34
Selenate [2]	71·67	79·94	91·09	8·27	19·42	2·35
Double Sulphate (Zn) [2]	196·16	205·58	217·97	9·42	21·81	2·32
Double Selenate (Zn) [2]	208·80	218·35	230·77	9·55	21·97	2·30
Perchlorate [3]	54·91	61·33	70·05	6·42	15·14	2·36
Permanganate [4] ...	58·53	63·23	70·01	4·70	11·48	2·44
$C_6H_4.COOH.COOM$ [4] ...	124·8	129·6	136·7	4·8	11·9	2·48
Metaperiodate [4] ...	63·7	70·6	—	6·9	—	—
Bichromate [4]	110·2	123·9	—	13·7	—	—

This constancy of the difference-ratio would seem to afford an exact numerical significance for the special term *eutropic*; thus in any isomorphous series derived from a given eutropic group of metals, it appears that the differences in molecular volume along the series stand in a constant ratio which is characteristic of the metals but is the same in all the series, and so is independent of the nature of the acid radicle. This relation is strictly confined to the eutropic elements,[5] and is not fulfilled by the salts of ammonium or thallium.

Calculation of atomic volumes.—Since the differences in all the isomorphous series of Table I stand in the same constant ratio, it follows that the volumes $V_1, V'_1, V''_1 \ldots$ of the salts [6] in any one series may be related to those in another series $V_2, V'_2, V''_2 \ldots$ by equations of the following type:

$$V_1 = aV_2 + C$$
$$V'_1 = aV'_2 + C$$
$$V''_1 = aV''_2 + C.$$

[1] See Landolt-Börnstein, Physikalisch-chemische Tabellen, 1920 edition.

[2] A. E. H. Tutton; see P. Groth, Chem. Kryst., vol. 2, pp. 323, 326, 490.

[3] A. E. H. Tutton, Proc. Roy. Soc. London, Ser. A, 1926, vol. 111, pp. 462–491. [Min. Abstr., vol. 3, p. 161.]

[4] Values taken from P. Groth, loc. cit.

[5] A. E. H. Tutton, Crystalline structure and chemical constitution, 1910, p. 125.

[6] The series in Table I relate to three metals only, but if more than three eutropic metals were available the relation would probably extend to them also.

For, by subtracting these equations in pairs, $V'_1 - V_1 = a(V'_2 - V_2)$ and $V''_1 - V_1 = a(V''_2 - V_2)$. Then, on dividing,

$$\frac{V' - V_1}{V''_1 - V_1} = \frac{V'_2 - V_2}{V''_2 - V_2},$$

this being the relation observed in the last column of Table I. C is a volume, constant for each isomorphous series, i. e. for each acid radicle, but independent of the metal. a is a factor, also constant, for the isomorphous series and independent of the metal; it is the ratio between any difference in the isomorphous series $V_1 \ldots$ and the corresponding difference in the other isomorphous series $V_2 \ldots$.

Now the metals themselves form one of these series (Table I); the volumes of the free metals may therefore be chosen as V_2, V'_2, $V''_2 \ldots$. Each of the three equations given above then assumes the form

$$V(\text{salt}) = aV(\text{metal}) + C.$$

Expressed in words, this implies that the volume of any salt in a given isomorphous (eutropic) series is the sum of a variable volume, which is a times the volume of the metal in the free state (if the free metals are isomorphous), and a constant volume C depending solely on the nature of the acid radicle. Reasons will be given below for supposing that the atoms in these crystals can be treated as relatively incompressible, but plastic, units completely filling the available space in the crystal; the above partial volumes will therefore be termed the partial volumes of the metal and acid radicle (or other residue) in the crystal. Values for these quantities are readily calculated from the data in Table I, and have been summarized in Table II. The results are derived from the average of

Table II. Calculated volumes of metal and acid radicle in salts of K, Rb, Cs.

	Metal (K).	Radicle.	Metal (Rb).	Radicle.	Metal (Cs).	Radicle.	Mean Radicle.
Sulphate	36·0	28·91	44·4	28·94	56·0	28·58	28·8
Selenate	35·5	36·17	43·7	36·24	55·1	35·99	36·1
Double Sulphate (Zn)	40·2	155·96	49·5	156·08	62·4	155·57	155·9
Double Selenate (Zn)	40·5	168·30	49·9	168·45	63·0	167·77	168·2
Perchlorate	27·6	27·31	34·0	27·33	42·8	27·25	27·3
Permanganate ...	20·8	37·7	25·6	37·6	32·3	37·7	37·7
$C_6H_4.COOH.COOM$	21·5	103·3	26·5	103·1	33·4	103·3	103·2
Metaperiodate ...	29·8	33·9	36·7	33·9	—	—	—
Bichromate	59·3	50·9	72·9	50·9	—	—	—

the ratios given by Rb–K and Cs–K. The calculation can also be made on any pair of salts for which isomorphism has been fully proved on

crystallographic grounds, and two examples of this class are included in Tables I and II.

B. *Series containing sodium and lithium.*

These salts are very rarely isomorphous with those of potassium, but several isomorphous pairs are available for sodium and lithium, and partial volumes can be calculated from the free metals by the method already used for potassium and rubidium. The values are given in Table III.

Table III. Molecular volumes of isomorphous salts of Li, Na.

Type.	Li.	Na.	Diff.	Calculated volumes. Li.	Na.	Radicle.
Metal [1]	13·0	23·7	10·7	—	—	—
LiCl [2]	20·39	26·72	6·33	7·69	14·02	12·7
LiBr [2]	24·90	31·97	7·07	8·59	15·66	16·3
LiI [2]	32·75	40·72	7·97	9·69	17·66	23·1
Li_3PO_4 [2]	48·1	64·7	16·6	20·15	36·75	27·95
$Li_2S_2O_6.2H_2O$ [2] ...	97·5	110·7	13·2	16·04	29·24	81·5
LiF [2]	11·3	16·4	5·1	6·2	11·3	5·1
Li_2O [3]	16·6	27·6	11·0	13·3	24·4	3·3
Li_2S [4]	27·9	41·95	14·05	17·07	31·12	10·8

C. *Isomorphous series derived from the halogens Cl, Br, I.*

Next to the alkali metals, these elements probably afford the best example of eutropic isomorphism. Data for a number of isomorphous series are collected in Table IV. Here the ratio (I–Cl)/(Br–Cl) is

Table IV. Molecular volumes of isomorphous salts of Cl, Br, I.

	Cl.	Br.	I.	Br–Cl.	I–Cl.	$\frac{\text{I–Cl}}{\text{Br–Cl}}$
K [2]	37·11	42·90	52·57	5·79	15·46	2·67
Rb	42·80	48·90	59·07	6·10	16·27	2·67
Cs	41·80	47·40	56·85	5·60	15·05	2·69
Li	20·39	24·90	32·75	4·51	12·36	2·74
Na	26·72	31·97	40·72	5·25	14·00	2·67
$Ba(X_2)$ [5]	54·1	62·2	76·0	8·1	21·9	2·70
$Ca(X_2)$ [5]	51·6	59·6	74·3	8·0	22·7	2·84

[1] See Landolt-Börnstein, Physikalisch-chemische Tabellen, 1920 edition.

[2] Values taken from P. Groth, loc. cit.

[3] J. W. Mellor, Treatise on inorganic chemistry.

[4] Values obtained from X-ray measurements, A. Claassen, Recueil Trav. Chim. Pays-Bas, 1925, vol. 44, pp. 790–794. [Min. Abstr., vol. 3, p. 245.]

[5] Landolt-Börnstein, loc. cit. No crystal measurements available.

constant in exactly the same way as the ratio (Cs–K)/(Rb–K) in Table I. The volumes will therefore be given by expressions of the type $V(\text{salt}) = a'V(\text{halogen}) + C'$, where a' and C' depend only on the metal, and V(halogen) is the volume the free halogen would have in an isomorphous series. The solid series is not known, but estimates of the relative volumes can be obtained from the salts of the alkali metals. These require more detailed discussion, for the values tabulated by Groth (Table Va) show a small but definite increment in the differences along the series.

Table Va. Molecular volumes of alkali halides.[1]

	Cl.	Diff. (Br–Cl).	Br.	Diff. (I–Br).	I.
K	37·11	5·79	42·90	9·67	52·57
(Rb–K) ...	5·69	—	6·00	—	6·50
Rb	42·80	6·10	48·90	10·17	59·07
Li	20·39	4·51	24·90	7·85	32·75
(Na–Li) ...	6·33	—	7·07	—	8·00
Na	26·72	5·25	31·97	8·75	40·72

It is evident that the formulae given do not hold good absolutely in these series; the 'constants' are apparently undergoing a progressive change. But this change could be eliminated if the data were observed under suitably graduated conditions of temperature and pressure; it would seem, therefore, that in this rather extreme case the 'corresponding states' should be defined even more closely than is done by taking the volumes of isomorphous salts at normal temperature and pressure.

So great an increment in the differences is exceptional; only a relatively small change occurs on the substitution of K, Rb, Cs, and of S, Se in the sulphates and selenates (Table I). It would seem, therefore, to be associated rather with the marked gradation of properties in chlorine, bromine, and iodine than with any peculiarity in the alkali metals; comparable conditions will therefore most probably be obtained by taking the volumes of the chlorides, bromides, and iodides under conditions of progressively greater compression chosen so as to eliminate the increase in the differences. This requires that the bromides of K, Rb should be compressed in the ratio of 6·00 / 5·69 and the iodides in the ratio 6·5 / 5·69. The result of this correction is shown in Table Vb. The differences are now substantially equal and the usual method of calcula-

[1] See P. Groth, Chemische Krystallographie, 1906, vol. 1, pp. 165, 166.

tion can be applied, yielding the partial volumes given in Table V c. A similar correction is required for the corresponding salts of sodium and lithium (also given in Table V), which then yield an independent estimate for the relative volumes of the halogens.

Table V b. Molecular volumes of alkali halides after compression of bromides and iodides.

	Cl.	Diff. (Br–Cl).	Br.	Diff. (I–Br).	I.
K	37·11	3·54	40·65	5·35	46·00
(Rb–K) ...	5·69	—	5·75	—	5·70
Rb	42·80	3·60	46·40	5·30	51·70
Li	20·39	1·91	22·30	3·63	25·93
(Na–Li) ...	6·33	—	6·31	—	6·30
Na	26·72	1·89	28·61	3·62	32·23

Table V c. Partial volumes in alkali halides (after compression).

	Metal.	Cl.	Br.	I.
K	24·65	12·46	16·00	21·35
Rb	30·35	—	—	—
Li	7·69	12·70	14·61	18·24
Na	14·02	—	—	—

This method of correction is, of course, only approximate, the general result being to minimize the increment in volume along the halogen series, but it may be of interest to compare various estimates for the relative volume of the halogens.

Relative volumes of the halogens.

	In Halides.			Elements.
	Richards.[1]	A. F. H.		
	Mean N. T. P.	Mean N. T. P.	Mean corr.	Observed values at boiling-point.[2]
Cl	13·3	12·4	12·6	21·7
Br	17·1	16·6	15·3	27·0
I	—	23·8	19·8	37·0

The apparent expansion of the bromides and iodides renders it difficult to employ the equations given at the beginning of this section to calculate partial volumes for the halogens in these salts. It would seem necessary in each case first to apply a correction for the change in pressure along the series; this unfortunately has a rather pronounced effect on the differences, so that the accuracy of the estimate of the size of the halogen

[1] T. W. Richards, Journ. Amer. Chem. Soc., 1923, vol. 45, pp. 422–437.

[2] See J. W. Mellor, Treatise on inorganic chemistry, 1922, vol. 2, pp. 50 and 66.

is adversely affected; only a rough estimate could be obtained by using the average values for the elements in the above table, and it seems better, when possible, to rely on the indirect estimate obtained by difference after calculating the volume occupied by the metal in the manner given for the alkalis.

General considerations.

The data here examined seem to indicate the operation of two general principles: first, that *in any two isomorphous salt-series formed from the same group of eutropic elements the volume of an element in one series bears to its volume in the other series a ratio which is the same for all members of the group*, and, second, *the eutropic replacement of one element by another of different volume does not alter the volume of the rest of the molecule.* It is very difficult to reconcile the close accuracy of these additive relations with the conception of the crystal as a structure of hard spheres; for there is no indication of a 'co-volume' which should represent the interstitial spaces. On the other hand, simple additive relations of this type would be entirely consistent with a structure built up of atoms which behave as if plastic and perfectly deformable under the stress of neighbouring atoms, but have only the small degree of compressibility common to the solid state. This mode of treatment, though perhaps somewhat at variance with the usual method of treating the atoms as hard spheres with interspaces, possesses certain advantages in dealing with other properties of crystals. For example, topic axes in an isomorphous group may vary in an apparently arbitrary manner while the volume is additive in accordance with the law of Retgers. This would imply that deformation of the lattice in two directions must always be compensated by an adjustment of the third axis, a result which lends itself to numerical treatment in accordance with the idea of a plastic atom. These considerations, though of a general character, seem to lend a measure of support to the view that the space-lattice might be treated as an aggregate of plastic atoms, of which the volume is relatively fixed; the volume of the atom itself being subject to variation with temperature and pressure, and also to discontinuous changes corresponding with the state of chemical combination.

Petrology of the Bodmin Moor granite (eastern part), Cornwall.

(With Plates VI and VII.)

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and Technology, London.

[Communicated by Prof. W. W. Watts, F.R.S. ; read June 14, 1927.]

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I. Introduction.

THIS paper presents the results of an investigation of the granite composing the eastern part of Bodmin Moor, Cornwall, as mapped on Sheet no. 337 of the Geological Survey. The work was undertaken on the suggestion of Dr. J. W. Evans, and comprised the field- and laboratory-work necessary for a detailed study of the petrology and tectonics of the area.

The boundary of the granite, as mapped by the Geological Survey, has been retained, and also the distribution of the two main granite types ('coarse' and 'fine') shown on the Survey map. The coarse granite, however, has been sub-divided into two varieties, and it has been found practicable to map the distribution of each (fig. 1).

The killas inclusions in this granite have not hitherto been studied, and these have received due attention. Similarly, the minor intrusions described include some that have not been previously recorded.

Bulk analyses of Bodmin Moor granites were lacking, and, as the investigation indicated the need of such analyses, representative types have been selected and analysed.

II. The Prevalent Rock-types.

(*a*) *The granite types and their distribution.*

The Survey map (Sheet no. 337) distinguishes between two different varieties of granite, namely (1) an earlier 'coarse-grained granite', and (2) a later 'fine-grained granite'.

The results of the author's investigation establish the composite character of the coarser granite, which presents two varieties differing in chemical composition and petrographical characters. One of these varieties corresponds in detail with the standard coarse-grained granite

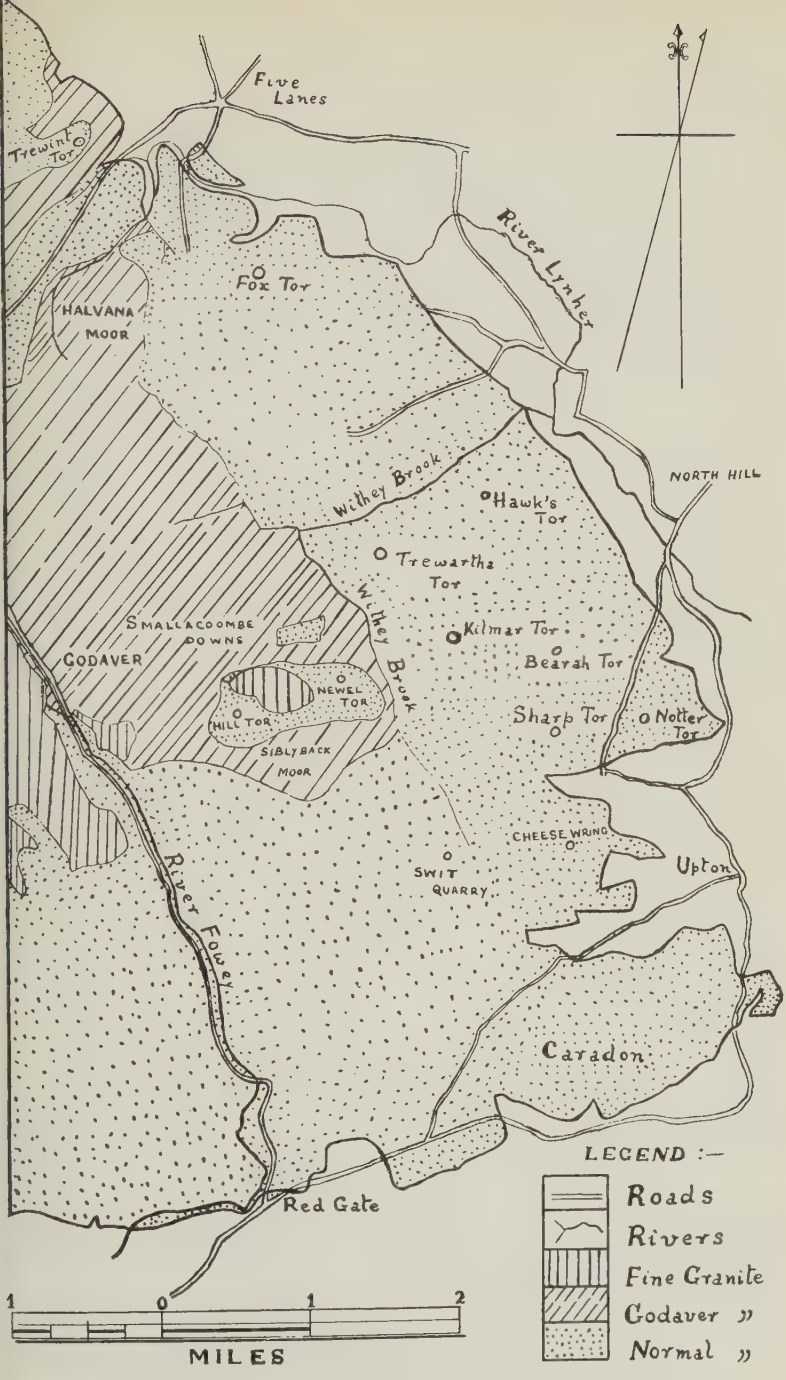


FIG. 1. Sketch-map of the Bodmin Moor granite (eastern part).

described in the Memoir, and will be referred to, herein, as the '*Normal type*'. The other variety, newer than the former, will be referred to as the '*Godaver type*'; the type-locality being in the region of the Godaver mines, just a little to the north of Harrow Bridge, east of the Fowey, where it was first noticed. The third type will be referred to as the '*fine type*', after the Geological Survey.

The distribution of the three types is shown on the map (fig. 1). The 'Normal type' predominates, forming the whole of the eastern and southern parts of the area and some portions on the north. It also forms some of the tor-caps,[1] resting on the 'Godaver type', in the region where that reaches its maximum development.

The 'Godaver type', with the exception of an occurrence west of the Fowey, bordering the east and north of the 'fine granite', is confined to the area between the Fowey and the Withey Brook and its northward extension. South of the typical area it appears on the flanks of the Smallcombe Tor, Newel Tor, and Hill Tor (all of which are capped by the 'Normal type'), and gradually falls in level towards the Siblyback Moor, where it eventually disappears. South of the main area it occurs again on the Craddoc Moor as dikes in the 'Normal' granite, e.g. in the Swit quarry.

North of the three tors named above, as far as the north side of Halvana Moor, the whole area is made up exclusively of the 'Godaver type'. On the north-east of Halvana Moor, the 'Godaver type' gradually dips beneath the Normal type, becoming finer-grained near the contact. On the slightly higher ground east of the small river, the Normal type is extensively injected by vein-like offshoots from the underground prolongations of the Godaver type beneath this area, and also by veins of quartz-tourmaline and quartz in large amounts. The Godaver type reappears again as a thin sill and vein injections in the Normal type in the region north of Fox Tor.

From the characters described above it would appear that the Godaver type was intruded almost horizontally below the Normal type. The contact-surface or 'roof' is practically horizontal towards the centre, e.g. Hill Tor, Trewint Tor, &c., but shows a gentle dip at the margins, e.g. Halvana Moor. Pseudo-bedding is parallel.

The character of the intrusive 'fine granite' has been described in Memoirs 336 and 337. It may be added that this also shows intrusive relations to the Godaver type at its eastern and northern margins, where

[1] Cf. the occurrence on Dartmoor described by Dr. A. Brammall, Proc. Geol. Assoc., 1926, vol. 37, pp. 262–264.

it sends vein-like offshoots and quartz-schorl veins into the Godaver type.

The Normal granite is laccolithic in relation to the country-rock, the pseudo-bedding showing a flat dip near the junction, while it is practically horizontal elsewhere.

Megascopic characters of the three types.—The Normal type is the coarsest in texture. Porphyritic orthoclase is abundant. Biotite and muscovite are present in almost equal proportions, and tourmaline also occurs. Quartz affords the most valuable guide to the identification of this type in the field; it occurs in large granular patches more or less interstitially.

The Godaver type is of medium grain and is also porphyritic, but its phenocrysts are less abundant than those of the Normal type. Pink orthoclase occasionally occurs in the groundmass. Biotite is less, and tourmaline more, abundant than in the Normal type. A little purple mica, lithionite, may be detected in hand-specimens. In contrast to the Normal type, quartz generally shows a porphyritic habit; it is more or less rounded, and does not appear granulated. The bleb-like character of the quartz is very characteristic. At its contact with the Normal type, the Godaver type has a finer texture, and may then resemble the 'fine granite'; but it never shows any great amount of chill-effects, and rapidly regains a medium coarseness at a short distance from the contact, where it is coarser than the 'fine granite' at its coarsest.

The 'fine granite', in addition to its fineness in grain, shows a few and very small porphyritic crystals of orthoclase, which sometimes vanish completely. 'The usual absence of the large quartz grains that are persistently present in the main intrusion . . .'[1] is also a remarkable feature. The amount of biotite is less and that of tourmaline more, than in the earlier types.

(*b*) *Minor intrusions and mineral veins.*

Each of these major igneous intrusions was followed by a suite of minor intrusions through fissures newly opened in them as they gradually cooled and consolidated. They occur in dikes, sills, and thin veins, and present many varieties, representative of an undifferentiated granite magma on the one hand, and its differentiated fractions, both leucocratic and melanocratic, on the other. In addition, there occur also mineral

[1] Geol. Survey Sheet Memoir, no. 337 (The geology of the country around Tavistock and Launceston), 1911, p. 70.

veins, e.g. quartz-tourmaline veins, and quartz-veins, joint-plane crusts of fluorite, and metalliferous lodes.

The intrusive history may be summarized thus:

1. Intrusion of the Normal granite, followed by quartz-tourmaline veins, and then by
   (a) Pegmatites and melanocratic veins,
   (b) Aplites,
   (c) Quartz-tourmaline veins, quartz veins.
2. Intrusion of the Godaver granite, followed by quartz-tourmaline and quartz veins.
3. Intrusion of the 'fine granite', followed by quartz-tourmaline and quartz veins.
4. Elvan dikes, followed by quartz-tourmaline and quartz veins.
5. Metalliferous lodes, followed by quartz-tourmaline and quartz veins.

(i) *Undifferentiated Type of Minor Intrusion.*—The rocks of the undifferentiated type, which are the commonest and occur chiefly as dikes, belong to the granite- and quartz-porphyry families, with the usual characters. They are locally known as 'elvans'. They follow the 'fine granite'. Their trend is roughly E.N.E. to E.-W., parallel with the mineral veins and lodes and to one of the major joints of the granite. A feature in connexion with many of these dikes is the large development of cubical crystals of pyrites, especially near the margins, embedded in the felsitic groundmass. Jointing is well-developed, often in three or more systems, which divide the rock into rhombohedral blocks.

(ii) *Differentiated Types.*—Turning to the differentiated types, we find pegmatites and highly felspathic veins (both of coarse texture), with aplites and melanocratic veins (both fine-grained), the latter very rich in biotite and plagioclase. The differentiated minor intrusions, with the exception of mineral veins and lodes, are associated with the Normal granite only and are pre-Godaver.

1. *Pegmatites and Felspathic veins.*

These occur generally as irregular and lenticular pocket-infillings of no great extent in the body of the main granite; the longest axis lies more or less on the horizontal plane. They are sometimes crossed by surfaces of the pseudo-bedding in continuity with those of the granite (e.g. at Notter Tor, upper quarry). They are very coarse-grained, with

large felspars, quartz, muscovite flakes, and tourmaline crystals. They sometimes occur as horizontal, sill-like sheets, and as narrow, more or less vertical dikes along the roughly east and west joint-planes of the granite. The rock of the dike is medium-grained, but coarser than the granite, that of the sill is coarser still. In some cases, the enclosing granite becomes mineralized near the contact.

2. *Melanocratic Differentiates.*

The melanocratic differentiates sometimes occur associated with the lenticular pocket-infillings of the pegmatitic material as thin films forming, in part at least, the external boundary of the pocket and then passing into the granite itself as a narrow and very irregular vein.[1]

Similar differentiates may also be found in the granite without any apparent connexion with the pegmatitic material, and show the same irregular habit. Thus, from a horizontal position along the pseudo-bedding of the granite, the rock may suddenly pass upwards through the granite in a deviating fashion with repeated minor veinings at all levels.

Small lenticular patches of this material may be found in the near neighbourhood of the veins, sometimes connected with them by minor veins. This connexion is not apparent in other cases, and the patches in consequence appear as a sort of segregation product. These are fine-textured, with porphyritic plagioclase and quartz and a large amount of biotite embedded in a bluish, even-grained groundmass. The biotite usually shows a banded arrangement. Andalusite is abundant.

3. *Aplites.*

These have been observed mainly in the Kilmar, Hawk's, and Trewartha Tors along the southerly-dipping E.N.E. joint-planes. A smaller number also occur in the other set of main joints. These are never

[1] Here perhaps we have an instance of rock-differentiation on a small scale. As the consolidation of the main body of the rock continued, there continued to collect in these pockets the remaining 'liquor', which had a high content of fluxes promoting crystallization. The liquor was, however, contaminated: andalusite, collected below and marginally, by interaction with the residual liquor, precipitated a large amount of plagioclase and biotite (see p. 304), and gave rise to the melanocratic rock (which still contains a large amount of andalusite); while the body of the fluid, being freed from the ferromagnesian and a large amount of more basic plagioclase content in this way, crystallized out as a perfectly leucocratic rock with orthoclase and albite as its dominant felspathic constituents.

thick, varying generally from 2 to 9 inches in breadth. They are always fine-textured and even-grained; they are light greyish-white in hand-specimens, showing a certain amount of light-coloured schorl and occasionally small flakes of muscovite. The aplites do not show any chilling at the margin, but, on the contrary, have been seen to grow a little coarser in some cases, though this marginal facies never attains the coarseness of a pegmatite, as in the Dartmoor granite.[1]

4. *Mineral Veins and Lodes.*

The mineral veins, viz. quartz-tourmaline, quartz, and fluorite veins, occur along the joint-planes of the granite and elvans, and also as abundant offshoots at the contact regions of the various granite types. They followed every one of the major and minor intrusions. The lodes appear as the last phase of igneous activity, and are found to cut through all the granites and elvans. These were also followed by quartz-tourmaline and quartz veins.

III. General Microscopical Character.

(a) *The Normal Granite Type.*—This type is very coarse-grained, with a large development of porphyritic crystals of microcline and orthoclase. None of the constituent minerals, except accessories, present well-defined crystal outlines. Even the porphyritic microcline and orthoclase show a rugged and resorbed margin, and there is almost always a narrow and irregular growth of plagioclase along the margin partly or almost completely surrounding the crystals. These plagioclase strips (albite, albite-oligoclase) may be composed of several small crystals; sometimes, individual crystals of fairly large size are also met with. Orthoclase and microcline also occur in the groundmass in irregular plates. The plagioclase is a little more idiomorphic, the smaller ones still more so. Coarse patches of granulitic quartz are usual and the boundaries are the least defined. The individual patches are composed of a large number of smaller individuals with interlocking and sutured boundaries.[2]

Biotite and muscovite are present almost in equal amounts, and are very often intergrown. Pleochroic haloes are abundant, especially in the former. Tourmaline is invariably present. Cassiterite, iron-ores (titaniferous or not), apatite, are very usual accessories; andalusite is

[1] A. Brammall, loc. cit.

[2] Cf. W. A. Richardson, A micrometric study of the St. Austell granite. Quart. Journ. Geol. Soc., 1923, vol. 79, p. 553.

a very characteristic accessory; corundum and zircon are not uncommon. These three minerals occur generally as inclusions in micas and felspars; but andalusite also occurs as an independent mineral. Topaz and fluorite are rarely present except in pneumatolysed varieties.

Sharp Tor—Notter Tor Sub-Type.—While predominance of potash-felspars over the plagioclase, and coarseness of grain of the groundmass in the Normal granite type, hold as a general rule, an exception in both these respects has been noticed in the case of the rocks of the Sharp Tor and Notter Tor areas. The mineral character and the texture, presenting several departures from the standard type, are noted below.

The potash-felspars are heavily charged with perthitic intergrowths. They also contain small inclusions of plagioclase as usual. The cross-hatching of microcline is on a very delicate scale and may disappear from a part of the crystal altogether.[1] The boundary of these felspars is exceedingly irregular and corroded, with large marginal deposition of plagioclase (albite–oligoclase). Some of the phenocrysts are irregularly cracked, often giving rise to wide interspaces (due evidently to melting under conditions of disturbed equilibrium), the infilling of which is exclusively plagioclase in subhedral plates, and rather narrow and elongated tablets. A considerable amount of the potash-felspars seems to have been removed in this way to make room for so much plagioclase; sometimes, only a very narrow strip of the original microcline is left as a relic among the growths of later plagioclase.

The smaller crystals of microcline and orthoclase in the groundmass also show the border zone of plagioclase—a fact never observed except in this type. These phenomena are perhaps somewhat akin to albitization, and there is, on the whole, a preponderance of plagioclase over potash-felspars. The proportion of $K_2O : Na_2O$ (4·26 : 4·31) when compared with that of the usual types (5·5 : 3·5, on the average), is also marked with a striking variation. (Vide Chemical Analyses 1, 2, & 3, p. 307).

While the rock is coarse-grained on the whole, there has been noticed a very delicate lace-like structure, in places as a groundmass, sometimes as a fringe to the larger constituents, and somewhat resembling 'myrmekite structure'.[2] In this case it consists, however, of slender rods and grains of clear orthoclase intergrown with vermicular and wart-like grains of quartz. Some of the muscovite plates ophitically

[1] This implies a large content of soda: J. P. Iddings, Rock Minerals, 1911, p. 235.

[2] J. J. Sederholm, Bull. Comm. Géol. Finlande, 1916, no. 48, p. 63.

enclose slender orthoclase laths. Shreds of biotite are also present in the groundmass; they belong to a later stage than large flakes—the usual habit of the mineral.

The rock presents another peculiarity; it is crossed by thin veins of clear orthoclase resembling in purity that in the granophyric groundmass. These traverse earlier formed potash-felspars and plagioclase and consist of a sub-granular aggregate of clear orthoclase crystals, with twinning rarely developed. In places, these are crowded with needles of blue tourmaline. Apatite is very often associated with these veins.

These rocks have been considerably injected by tourmaline veins towards the later stages of consolidation, and in connexion with them there has been a development of small needles and grains of tourmaline in the granophyric groundmass. Some sections, cut perpendicular to the vein, show under the microscope the development of tourmaline on one side of the vein only, while the other half of the rock is quite unaffected.

(i) *Microscopical Characters of Individual Minerals.*

Microcline is generally fresh and clear as opposed to plagioclase. Carlsbad-twinning is often present in addition to the usual cross-hatching. The latter type of twinning is sometimes on a very delicate scale and may disappear from a part of the crystal altogether. The porphyritic felspars specially are largely perthitic. There are also, in addition, inclusions of twinned and zoned plagioclase crystals, generally of tabular habit with good crystal outlines; also of elongated narrow prisms, sometimes along the composition-plane of the twin itself. These may also exhibit a tendency to occur zonally; in a few observed cases, four or five alternate zones could be made out. Thus in one case, the core is formed of a Carlsbad-twin of microcline, followed and surrounded on all sides by a narrow zone of small crystals of plagioclase; then follows another wider zone of microcline in reversed orientation relative to the core. This is again followed by a less definite zone of plagioclase, and then by microcline in reversed orientation relative to the nearest zone (Plate VI. fig. 1). The later zones are very much interrupted by the intergrowth of plagioclase with irregular boundaries and large flakes of mica (muscovite), some of the latter with inclusions of microcline in optical continuity with the phenocryst of microcline itself. Some of the included plagioclases may show neither zoning nor twinning, but are marked by undulatory extinction (?anorthoclase). The included plagioclases themselves may carry inclusions of idiomorphic biotite.

Inclusions of quartz are also present. They may be quite irregular in shape, occurring throughout the whole mass of the crystal; or they may occur as numerous rounded grains at the border zone and optically continuous among themselves.

Phenocrysts are often cracked and filled with later minerals. There is often a zone of spongy and fibrous muscovite fringing the contact of the phenocrysts with plates of muscovite (Plate VI, fig. 1).

Orthoclase is present in far greater abundance than microcline and also presents the above characters.

Plagioclase is generally turbid with decomposition products. When zoned, the centre is always more decomposed than the exterior. Even when present as inclusions in the porphyritic potash-felspars, these characters seldom fail. The results of the cleavage fragment tests, supplemented by the extinction-angle method are recorded below:

Relative *n* of felspar fragments.	Medium used.	Orientation.	Ext. angle.	Composition.	Remarks.
(i) *n*	>Clove oil	(010)	−9°	$Ab_{59}An_{41}$	Not very frequent.
(ii) *n*.	Clove Oil >Chlor-benzene	(010) and (001)	0°	$Ab_{70}An_{30}$	Frequent.
(iii) *n*	<Clove Oil >Chlor-benzene	(010)	+(12–15)°	Albite-Oligoclase	Very frequent.

Thus among the plagioclases, there is a preponderance of oligoclase over albite, the latter occurring generally as a marginal zone to the more basic species, oligoclase-medium andesine ($Ab_{59}An_{41}$). The preponderance of oligoclase has been noted by Flett as one of the special and distinguishing features of the Bodmin Moor granites, the felspars of other Cornish granites being generally of a more acid type.[1]

Plagioclase may carry inclusions of other early minerals in addition to quartz grains. The latter sometimes occur as innumerable rounded blebs at the periphery, and these blebs are in optical continuity among themselves.

Micas and Tourmaline. What has already been noted about muscovite and tourmaline in the St. Austell granite by Flett[2] and Richardson[3], regarding their long continued period of crystallization, applies very largely to biotite also in this part of Bodmin Moor. This phenomenon has been referred to the action of gases such as hydrogen, fluorine, and boric acid.[2]

Thus as inclusions, biotite may carry not only the earlier minerals (ores, apatite, and secondary rutile in altered varieties) but also felspar and quartz. Sometimes, a single grain of apatite may serve as a nucleus for both mica and plagioclase, being partially enclosed in both; also when micas segregate, crystals of plagioclase may serve as nuclei. Small but idiomorphic crystals of biotite occur as inclusions in plagioclase. These facts suggest that biotite started to crystallize before the plagioclase, but that its crystallization-period covered that of plagioclase and orthoclase and overlapped that of quartz.

Quartz generally occurs in patches of rather large size, and being as a rule the last product of crystallization, idiomorphism is practically wanting. These large patches, however (as noted by Richardson in some varieties of St. Austell granites), are resolved microscopically into aggregates of smaller crystals interlocked with one another along sutured boundaries. Undulose extinction and cataclastic structures are a general feature. There is often a development of two systems of cracks roughly at right angles to each other, noted also by Richardson and referred by him to imperfect rhombohedral cleavage of the

[1] J. S. Flett, Geol. Survey Sheet Memoirs, 335; 336, p. 54.
[2] J. S. Flett, Geol. Survey Sheet Memoir, 347, pp. 58, 59.
[3] W. A. Richardson, Quart. Journ. Geol. Soc., 1923, vol. 79, p. 565.

mineral. Sometimes one set only may be developed; when viewed under crossed nicols, extremely fine twin-like striations may sometimes be observed. This feature has been noticed also by Franchi[1] in the Ligurian granites, Italy. According to Rosenbusch[2] these may perhaps be twin-lamellae produced by strain.

Accessory minerals will be referred to in a later section (p. 307).

(ii) *Order of Crystallization.*—With slight variations, the sequence described by Flett in the case of the minerals of the St. Austell granite holds good for this area also. Here apatite seems to be distinctly later than zircon and ores, as the latter are found to be included in the former either partially or completely. Biotite in the St. Austell granite had a more or less definite period of crystallization, but here it falls in the same category as tourmaline and muscovite: the two latter exhibit the same tendencies in both places regarding their overlapping periods of crystallization. Felspars and quartz belong very largely to the normal order of appearance. Summarizing, the sequence of crystallization appears to have been as follows:—Ores and Zircon; Apatite; Andalusite; Biotite and Tourmaline; Muscovite; Plagioclase; Orthoclase and Microcline; Quartz.

(*b*) *The 'Godaver Granite' Type.*—While the mineral assemblage in this type is practically the same as in the 'Normal' type, and in a general way reproduces the same features under the microscope, there are a few distinguishing features worth noting:

(1) The proportion of biotite is decidedly smaller, and

(2) A little lithionite mica, purple-brown to light purple in colour, is present.

(3) There is a larger development of tourmaline.

(4) The plagioclase is more acid, not exceeding oligoclase in basicity. The oligoclase occurs as small cores, often cloudy in appearance, surrounded by a clear zone of albite.

(5) Quartz presents the most important variation. It has all the aspects of a porphyritic crystal. It forms large crystals more or less rounded at the margin and scarcely presents the appearance of the large, ill-defined, patches of granular quartz of the Normal type. The quartz of the second generation is not idiomorphic. It may occur as an idiomorphic inclusion in felspars.

(6) The rock is also finer in grain than the Normal granite.

In addition to the usual type, there are certain sill-like intrusions into the Normal type, which have been regarded as offshoots of the Godaver granite. These may be conveniently dealt with in this connexion.

They are granite-porphyries, much finer in texture than the typical Godaver granite, but they never show a chilled or felsitic selvage even at the contact, as the elvans and dikes of later ages so often do. At the

[1] S. Franchi, Boll. R. Com. Geol. Italia, 1893, no. 43.

[2] H. Rosenbusch, Mikroskopische Physiographie, 1907, vol. 2, p. 41.

actual contact, they are a little finer but regain their medium texture within a very short distance.

They contain porphyritic microcline, orthoclase, and quartz as in the main mass; and muscovite and biotite showing the usual characters. Tourmaline of usual habit is present; in addition, a large development of tourmaline occurs as grains and needles in the groundmass. They show a fair development of topaz, an unusual mineral in their plutonic representatives. This is perhaps connected with 'auto-pneumatolysis'.

(c) *The 'Fine Granite'*.—In this type, the amount of porphyritic orthoclase has markedly diminished, and microcline is very rare. The texture is much finer. The general groundmass is hypidiomorphic and more or less equigranular—excepting small local developments of slender quartz and orthoclase in fine granophyric intergrowths. A few peculiarities are noted below:

Orthoclase. The phenocrysts show perthitic intergrowths, which, however, vary considerably in thickness from place to place. Groundmass orthoclase is not perthitic.

Plagioclase diminishes in basicity. Oligoclase is much smaller in amount, the average extinction on (010) cleavage fragments varies from $+11°$ to $+14°$; the composition is between $Ab_{85}An_{15}$–$Ab_{90}An_{10}$. The plagioclase tablets are often notched by numerous irregular growths of quartz of elongated and bulging shape. These may be confined to a small part of the border zone only, or may penetrate far towards the interior of the crystal, leaving only a small core unattacked. These quartz grains are optically continuous and resemble a granophyric intergrowth.

Quartz. Crystals are never large, as in the first two types of granite. It may nevertheless show cataclastic structures and compose a rough mosaic.

Tourmaline is much larger in amount than in the earlier types; in common with that of other constituents, its coarseness also diminishes. In contrast with tourmaline of the earlier types, it tends to be idiomorphic, and is earlier than biotite, being found as idiomorphic inclusions in the latter and in muscovite. It may also occur as an inclusion in felspars, and partially in topaz also.

Biotite and Muscovite. The former is smaller in amount than in the earlier types. The two minerals present the same features as in the main intrusions. In the case of biotite, the overlap in the period of crystallization is even more emphasized than before. Flakes of biotite may occur as a sort of matrix in the interspaces between crystals of quartz and felspar. It may carry inclusions of quartz. In muscovite, both quartz and orthoclase occur as inclusions.

Topaz has not been found as a normal constituent in the earlier types. Here, however, its occurrence is very common. It occurs as flat, rectangular tablets, partially or almost completely enclosing idiomorphic tourmaline and presenting quite a sharp boundary to felspar and quartz. Prism-faces are well-developed; cleavage is not very sharp.

Andalusite is very small in amount and occurs as an inclusion only. It had hitherto been regarded as absent from this particular rock. Heavy residues, however, yield the mineral. This marked diminution of andalusite is a departure from the usual character of the rocks of Bodmin Moor.

In connexion with the diminution of andalusite and the corresponding frequency of topaz with a consequent higher fluorine-content, as borne out by chemical analysis as well, it is interesting to recall Flett's contention regarding the relationship of the two minerals.[1]

(*d*) *Aplite Vein.*—Typically intergranular in texture, and composed of rectangular plagioclase tablets, often meeting at an angle, the angular interspaces being composed of granular quartz, irregularly formed felspars, and other constituents. Orthoclase is also present and may carry inclusions of quartz. Quartz is irregular in outline and often shows cataclastic structures, specially marginally. Both biotite and muscovite are present, usually as an intergrowth of very small flakes. They are very subordinate. Tourmaline, in subordinate amount, occurs as grains and slender rods. When included in quartz and felspar, it is greenish; in felspars, such inclusions are often arranged parallel to their elongation. When not an inclusion, the tourmaline forms yellow-brown crystals, with or without blue cores, the crystals being stouter and idiomorphism less marked.

(*e*) *Melanocratic Differentiate.*—The texture is porphyritic, the porphyritic constituents being:

Quartz. Outline rounded; often cracked or broken up; inclusions of biotite present.

Plagioclase. Large, zoned crystals with altered cores and inclusions of quartz, biotite, &c. Twinning and cleavage ill-developed. The groundmass plagioclase may be twinned; zoning is not well-developed, but cleavage is usually distinct. The crystals are smaller in size but more numerous than those of orthoclase. The latter is very fresh, and untwinned; cleavage is distinct.

Biotite. In large jagged flakes showing numerous pleochroic haloes. It may tend to form aggregates, or may be intergrown with muscovite. It never shows rectilinear outlines, and it encloses quartz grains, felspar, &c., partly or completely. The biotite is more abundant than in the granites.

Muscovite. Small in amount but forming large flakes intergrown with biotite. When ending against orthoclase, the flakes are fringed.

Predominating accessory minerals are:

Andalusite. Plentiful; as grains or laths, marginally altering to white mica. Marginal zones are colourless; central zones violet or colourless.

Iron-ores.

IV. Altered Facies of the Granites.

(*a*) *Hydrothermal and Pneumatolytic alteration of the Granites.*—Altered facies of the granites have been observed chiefly with mineralization and with the intrusion of later into earlier granite-types. Three main phases are distinguished:

1. *Hydrothermal*—generally connected with the formation of tin-bearing veins.

[1] Geol. Survey Sheet Memoir, 347, p. 58.

2. *Fluoritization and Topasization*—with the development of secondary mica, fluorite, and topaz.

3. *Tourmalinization.*

The three phases generally occur together, the different pneumatolytic agents having apparently acted through the same channels. Thus cassiterite, fluorite, topaz, and tourmaline may be associated together in the altered granite—the first being, however, of somewhat restricted occurrence.

A few cases have been observed in which alteration, though referred to one or other of these alteration-processes, has yielded the products of the other processes in very small smounts. Such cases serve to indicate the sequence of the processes listed above.

Hydrothermal Phases.—Slide 179 shows the characteristic features: Slight development of secondary tourmaline and quartz; the clouding of felspars with opaque decomposition products—plagioclase being much more affected than orthoclase and often stained with chloritic matter. Biotite passes into radiating fan-shaped aggregates of green chlorite, often zoned, the centre being brownish, intimately associated with cassiterite. Cassiterite may be granular and brownish-red, or may show a rectangular habit, and may be zoned. Long prisms of steel-grey colour have also been noticed. It may also occur as small needles included in secondary quartz and arranged parallel to the dihexahedral prism faces. Apatite is often associated with the chlorite. Secondary tourmaline may surround cassiterite and may 'eat' its way into biotite.

Fluoritization and Topasization. In connexion with the development of topaz, felspar is highly sericitized, and biotite is bleached. Topaz may be attended by a little cassiterite and fan-shaped aggregates of bleached biotite. Tourmaline may be present, surrounding grains of topaz and cassiterite. During fluoritization, biotite does not seem to be affected, nor is felspar. When topaz develops in addition to fluorite, both biotite and felspars are altered. It is possible that this mode of alteration of biotite during topasization may be due to the action of fluorine in the presence of the (—OH) radicle, and that fluorine has no such potency when acting alone, as in the development of fluorite.

Tourmalinization does not alter either the form or colour of biotite, but often eats into it as well as into the plagioclase-felspars. Secondary quartz is almost always associated with this type of change, and carries abundant needles and grains of tourmaline. This type of alteration is generally confined to very narrow zones of the granite alongside the tourmaline-vein itself. The first two modes of alteration usually penetrate a little farther into the interior of the granite, as in similar occurrences in Scilly, where '. . . peach-veins are often seen to be fringed with a thin band of granite altered to greisen.'[1]

(*b*) *Pneumatolysis—a contact phenomenon associated with the later intrusive members of the sequence.*—An earlier granite at its contact with either of the later types shows fan-shaped bleached biotite-aggregates,

[1] Geol. Survey Sheet Mem. 335, 336, p. 57.

and large crystals of secondary topaz associated with sericitized felspar. The later granite shows these peculiarities and, in addition, a large development of idiomorphic groundmass-tourmaline, forming a matrix surrounding the phenocrysts of quartz and felspar. The amount of tourmaline is much larger near the contact.

Though the earlier rock has been affected by fluorine (with topasization as a result) it is quite unaffected by tourmalinization: it shows no fresh development of tourmaline. The later granite near the junction is traversed by thin veins of quartz, sometimes containing small schorl needles. These veins cut through all the earlier crystals and are more or less at right angles to the contact-surface of the two granites. These phenomena are very well seen in thin sections across the actual junctions of the two rocks.

(*c*) *Superficial Alteration.*—Agencies of weathering have acted on the granite producing wide-spread decomposition and kaolinization, superficially. The deep and workable deposits of china-clay, which are of limited occurrence in this area, are probably attributable to the action of carbonic acid and water emitted during the paulo-post stage of consolidation of the granite.

In this connexion, may be mentioned the alteration of granite beneath the high-level peat on the uplands. Under the peat occurs a thin layer of white soil about an inch in thickness, passing downwards into slightly disintegrated granite and thence into the solid rock. Analysis of a sample of this disintegrated granite taken beneath its soil-cover under 9 inches of peat, shows that the Fe_2O_3-content is reduced to an equivalent of FeO, with a little gain in moisture, a trace of CO_2 also being detected (vide chemical snalysis, p. 307). This may be a distinct kind of alteration, having nothing to do with the production of extensive kaolin-deposits, for Blanck and Rieser[1] have shown that kaolin does not result from the decomposition of granite under peat.

V. Inclusions.

The following table shows the various types of inclusions, classified according to their modes of origin and present distribution:

A. *Sedimentary*:

1. Hornfelses in Normal and Godaver granites; plentiful in the Normal granite.

[1] E. Blanck and A. Rieser, Chemie der Erde, 1925, vol. 2, 15. [Min. Abstr., vol. 3, p. 41.]

B. *Igneous*:

2. Melanocratic differentiates in the Normal granite.
3. Leucocratic differentiates, highly felspathic, in the Normal granite.
4. Micaceous segregations, in ovoid spots, around apatite and corundum grains.
5. Xenocrysts (pegmatitic felspars only) of Normal granite in dikes of Godaver granite.
6. Blocks of Normal in Godaver and Fine granites.
7. Fragments of No. 2 (above) in Godaver granite.

Although all the above types have been studied, the sedimentary xenoliths (no. 1, above) alone will be dealt with in detail in the following pages, as they present the most interesting features.

Hornfelsed (sedimentary) Inclusions.

(*a*) *Megascopic Character.*—Hornfelses occur throughout the granites, specially in the Normal type. In general, they are dark-brown in colour, and are of finer texture than the granite. The usual colour is of course due to the preponderance of biotite. In hand-specimens they may present a schistose appearance—alternate bands being made up of biotite on the one hand, and quartz and felspars on the other. Purple andalusite, though subordinate in amount, is also recognizable in the common types.

In addition to the prevalent type (biotite-hornfels) other varieties also occur, e.g., inclusions which are purple in colour and are made up chiefly of andalusite; and greenish-blue aggregates of sillimanite and corundum. Such types are, however, very scarce.

Bridging the gap between these extreme types, occur inclusions of hornfelsed killas, rich in biotite enclosing porphyroblastic sillimanite and corundum or andalusite in a lenticular fashion. With a gradual increase in the amount of biotite and a decrease in other constituents, these pass into the normal biotite-hornfelses—the predominant types.

Inclusions of the normal biotite-hornfelses are ovoid and variable in size, though usually small (2 to 2·25 cm. along the greatest diameter). Exceptionally large ones are of rare occurrence: the largest yet found has a diameter of 12 cm. In the granite, immediately margining these inclusions, are distributed still smaller fragments of the same material, which point to the shattering and dispersal of relatively large masses by the magma. Microscopical examination of the granite samples taken at, and near, the junction of granite with inclusions always reveals such

fragments in all stages of disruption, till finally only a microscopic fragment is left intact.

These inclusions end abruptly against the granite, and anything like an intermediate or transitional zone is rarely seen. An interesting variation, however, occurs in connexion with a specially large inclusion of biotite-hornfels: a rim of potash-felspar, about 1 cm. thick, is moulded on to the inclusion itself, ending abruptly against the granite; the rim only partially surrounds the inclusion; the ferromagnesian minerals of the granite, at the junction with the rim, are crowded together and arranged parallel to the contact; in no case does the granite show any 'chill-effect' or difference in texture at contact with the inclusion.

As regards distribution, the different types of inclusions show no preference for any particular depth-zone or peripheral zone. All the varieties may be found closely associated in one and the same sheet of granite.

(*b*) *Microscopical Character.*—For convenience of description and discussion of their microscopical character, the hornfelses are grouped as follows:

1. Biotite-hornfels type.
2. Andalusite-spinel-cordierite type (with subordinate corundum and biotite).
3. Sillimanite + andalusite-corundum-spinel type (with much biotite).
4. Sillimanite + andalusite-spinel-quartz type (with much biotite).
5. Andalusite-quartz-felspar type.

1. *Biotite hornfelses.*—These are generally schistose, specially near the contact with the granite (Plate VI, fig. 2). Far in the interior, the texture becomes more open, and is sub-ophitic to intergranular, with lath-shaped biotite embedded in a groundmass of granular quartz and plagioclase.

At the schistose margin, the planes of schistosity are constituted by alternating bands of dark-brown biotite and light-coloured minerals, chiefly felspars. In these bands biotite, specially, shows very close packing; individual crystals may be distributed at all angles to one another, but generally display a sub-parallel arrangement.

A large amount of magnetite often occurs in the biotite zone. Other minerals associated with biotite are, a little brown tourmaline (derived from the biotite itself), very little muscovite (larger in size than biotite and sometimes intergrown with it), grains of colourless to bluish corundum (which may show pseudo-cleavage), apatite, and a little quartz. Biotite contains pleochroic haloes around inclusions of zircon, and shows well-developed crystal outlines. Farther inward, however, schistosity is less definite and there is a large influx of felspar and quartz, and the biotite shows a jagged appearance (fig. 6). It may sometimes be found altering to titaniferous minerals, the latter associated with violet fluorite.

In the colourless bands, the chief mineral is plagioclase as small grains and tablets, ranging from albite-oligoclase to medium andesine; it is zoned, the centre being of higher refractive index and turbid; it shows scarcely any twinning. A few sporadic large crystals have also been noticed, surrounded by biotite in a lenticular fashion.

Other minerals are very subordinate: orthoclase (generally untwinned and fresh, sometimes with inclusions of quartz) and quartz, as small granules, sometimes with inclusions of biotite and tourmaline.

Injection veins of quartz also occur, cutting the planes of schistosity of the inclusion at an angle (Slide 172). The component grains of quartz are larger than those first described, and are often fractured. The veins represent injections from the magma.

More remote from the granite, schistosity disappears, and felspars, especially orthoclase, tend to become smaller in size. This is evidently connected with a larger supply of the requisite components near the granite-contact. The marginal schistosity may also be due to the pressure from the granite, probably the pressure set up at the time of crystallization of the surrounding magma.

As has been already noted, the texture becomes intergranular to sub-ophitic in the interior of the inclusion. Another remarkable feature is that, andalusite, often in very large flakes, occurs only in the interior of the inclusion, while the portion of the inclusion near the granite is perfectly free from it. The crystals of andalusite in the interior are surrounded by a zone of clear and colourless secondary muscovite, followed by zones of biotite and plagioclase (fig. 6).

All the constituents, except andalusite, are very much smaller in size than those in the granite. But the xenolithic andalusite is much larger in size than that in the granite, and resembles the porphyroblastic mineral of typically contact-altered aluminous rocks of this area.

In its mineral assemblage and texture, this type of inclusion most nearly approaches an igneous rock.

Rim of potash-felspar round an inclusion (Slide 172).—This consists of a multiple aggregate of potash-felspars (showing faint cross-hatching, perthitic structures and Carlsbad-twinning), growing radially outwards from the contact of the inclusion towards the granite. These felspars contain inclusions of small rounded blebs of quartz and small crystals of plagioclase; the former increase in amount as the inclusion is approached, and at the contact they are innumerable, and the microcline in consequence has a very jagged appearance; the plagioclase crystals, on the other hand, are scanty and may almost disappear near the contact with the inclusion, though steadily rising in amount towards the granite contact. Biotite, of a slender habit and in scanty amount, may also be included in this rim of microcline (figs. 4 and 5).

The granite at the contact with this outgrowing felspar-rim shows biotite crystals arranged with their long axes parallel to the contact (fig. 4). This has evidently been brought about by the growing pressure of the felspar itself as it crystallized outwards towards the granite magma.

(i) *Origin of the Biotite-hornfelses.*—Although texture and mineral assemblages (apart from andalusite and corundum) are those of an igneous rock, a possible igneous origin may be at once dismissed on the basis of chemical analysis alone (vide analyses, nos. 7 and 8, p. 307). The parent-rock was evidently shaly, and highly aluminous, as is indicated by the high percentage of Al_2O_3, and the

presence of andalusite, corundum, &c. The aureole of sedimentary rocks is made up of shales of various kinds.

Regarding the way in which this alteration of the shales was brought about, the idea of melting and re-crystallization can at once be dispensed with: according to Bowen, any change that could be brought about would be only by 'reaction-precipitation'.[1]

Attention will be confined to the alumina content only (as being the most important constituent of the parent-rock), its behaviour under conditions of metamorphism being well understood.

Dr. Evans[2] suggested that in the case of a magma reacting on aluminous sediments, anorthite and rhombic pyroxene are precipitated, e. g. the charnockite series in southern India. Bowen accepts this view in its entirety in the case of basic magmas, and thinks that in the case of acid magmas we should rather get micas than pyroxenes.[3] When a magma is particularly rich in volatile fluxes, as this granite magma was, we should perhaps expect the formation of micas in place of pyroxene.

In these Bodmin Moor inclusions, the aluminous matter re-acting with the granite magma precipitated a very large amount of plagioclase and biotite (and incidentally a little orthoclase and muscovite). The formation of andalusite and other minerals characteristic of contact-metamorphic rocks may be regarded as 'intermediate steps' in the process of alteration of the original shale. In these inclusions, andalusite is characteristically absent near the granite contact, the reaction evidently having proceeded to completion there, while its sporadic presence in the interior suggests its survival because of the exhaustion of the liquid magma.

(ii) *Formation of the felspathic rim.*—In the process of precipitation of plagioclase and biotite, the magma, in the immediate neighbourhood of these inclusions, became more or less depleted of ferromagnesian-, lime-, and soda-content, relatively to potash. The rim of potash-felspar may thus represent the material left over by the reaction. In this connexion the dearth, in this rim, of plagioclase inclusions but abundance of quartz near the contact with the hornfels, and the exactly opposite phenomena near the contact with the granite, is suggestive. The felspathic liquid, now represented by the rim, under favourable circumstances started crystallizing at the very place of its origin, i.e. just along the margin of the inclusion and grew outwards towards the granite, and during its growth outward, naturally pushed the biotite crystals of the granite magma in a line perpendicular to the direction of its growth. This may explain the 'packing' of the biotite, parallel to the line of contact with the inclusion.

The two analyses of inclusions of this type (nos. 7 and 8, p. 307) are, possibly by chance, remarkably uniform. The two samples were collected from different localties and from different situations; one was an inclusion in the Normal granite, the other an inclusion in a dike-modification of the Godaver granite. The original rocks were evidently of the same type, and their modification by granite had gone on almost to the same extent.

[1] N. L. Bowen, The behaviour of inclusions in igneous magmas. Journ. Geol., 1922, vol. 30, pp. 513–570.

[2] J. W. Evans, Quart. Journ. Geol. Soc., 1921, vol. 77, p. 133 (discussion on Tilley's paper).

[3] N. L. Bowen, loc. cit.

2. *Andalusite-spinel-cordierite type* (with a little corundum and some biotite).—Slide 44 a shows large plates of purple-coloured andalusite, with films and narrow elongated flakes of biotite intergrown along the prismatic cleavages, containing numerous inclusions of dark, opaque grains (graphite ?), and irregular grains of green spinel, sometimes decomposing marginally into fibrolite. These are enclosed as lenticules in an outer zone of small biotite crystals (fig. 8). Cordierite, as large crystals of tabular and rectangular habit, often pinitized, is arranged more or less parallel to the andalusite, with a partially intervening zone of quartz (fig. 7). Well-shaped corundum crystals occur in small amount in association with biotite and andalusite. A little muscovite and quartz are present interstitially, and a small amount of felspar occurs near the granite contact. Here appear four phases in a three-component system—a departure from the 'mineralogical phase rule' established by V. M. Goldschmidt, e. g. andalusite, corundum, cordierite, and spinel. This does not necessarily indicate 'a failure of equilibrium' as Tilley appears to consider [1]; according to Bowen, 'they could occur throughout a range of conditions rather than merely at a point, and there is no reason why these conditions might not be met with in nature.[2]

3. *Andalusite + sillimanite-corundum-spinel type* (with much biotite).—Slide 185 shows large stout plates of sillimanite, often in parallel intergrowth with acicular and platy andalusite. Large crystals of corundum with three rhombohedral pseudo-cleavages well developed, and partially filled with decomposition-products; the mineral often shows graphic intergrowth with biotite at the contact, and also penetration by needles of sillimanite. Its intergrowth with a sillimanite plate is also noted (figs. 9 and 10). Quartz and magnetite may be present as inclusions. Green spinel grains associated with sillimanite and corundum. The porphyroblastic constituents are surrounded by a matrix of biotite mainly, and a little quartz. Tourmalinization of biotite is usual. Corundum and andalusite also show tourmalinization, many relics of these altering minerals having been found in all stages of attack—some occurring at the centre of the tourmaline crystal.

4. *Sillimanite + andalusite-spinel-quartz type* (with much biotite).—Slide 178 shows a plexus of biotite with pleochoic haloes, surrounding, and very often intergrown longitudinally with sillimanite, the latter often in divergent needles with a little quartz in the interspaces. Sillimanite is often decomposing to fibrolite. Andalusite, very often in longitudinal tablets, and grains, altering to micaceous substances. It may sometimes show parallel intergrowth with needles of sillimanite. It also occurs as inclusions in micas. A little green spinel, embedded in andalusite and sillimanite. Tourmalinized biotite, with the original sillimanite-intergrowth still preserved (fig. 11).

Groups 2, 3, and 4 of the hornfelsed inclusions include types which have the mineral-assemblages produced in contact-metamorphism of the system MgO-Al_2O_3-SiO_2.[3] These have been modified very slightly, or not at all, by the

[1] C. E. Tilley, Quart. Journ. Geol. Soc., 1924, vol. 80, pp. 68, 69.

[2] N. L. Bowen, Journ. Washington Acad. Sci., 1925, vol. 15, p. 280. [Min. Abstr., vol. 3, p. 88.]

[3] C. E. Tilley, Paragenesis of the minerals of the three component system MgO–Al_2O_3–SiO_2 in thermal metamorphism. Geol. Mag., 1923, vol. 60, pp. 101–107.

granitic magma (with which equilibrium was not attained) and may be regarded as 'intermediate products', surviving only because of the exhaustion of the liquid.

These present a marked contrast in texture to the biotite-hornfelses. The habit of the constituent minerals is truly porphyroblastic, generally with an insignificant matrix of biotite and quartz enclosing the large crystals of andalusite, sillimanite, and corundum in a lenticular manner.

A certain peculiarity of behaviour of cordierite is also noticeable in these types. Cordierite arises when there is a very slight or no development of biotite (type 2); but it is absent in types which are very largely biotitic (types 3 and 4). This is also more or less the case in contact-altered shales, the presence of cordierite being connected with a limiting value of FeO-content in relation to MgO. To quote Tilley (loc. cit.):

'With a high FeO-content it is possible that cordierite may disappear, and it is of interest to note that available analyses of rock-forming cordierites show a limiting value of FeO-content.'

Thus in types 3 and 4, the FeO-content having perhaps exceeded the limit, as is suggested by the large amount of biotite, cordierite has been entirely suppressed.

5. *Andalusite-quartz-felspar type.*—Andalusite, as large, tabular plates of purple colour, sometimes zoned (purple centrally, colourless at the margin); or as anhedral grains and elongated, lenticular forms, as well as lozenge-shaped rhombic sections and cruciform aggregates, the latter embedded in large, ophitic or sub-ophitic quartz which may also enclose small tablets of plagioclase. Plagioclase may also enclose andalusite. Quartz shows undulatory extinction and is often fractured. The plagioclase is albite-oligoclase to oligoclase, and more or less turbid. There are also wedge-shaped injection-veins of later quartz cutting through some of the rock-minerals. Near the granite-contact, quartz and felspar tend to become larger in size. No orthoclase and no mica occurs (fig. 12).

This type is quite different from the others already described in having no mica. In texture, it closely resembles an igneous rock, and hence is quite unlike types 2, 3, and 4. This has evidently been affected by the granite magma, more or less like type 1, which it most resembles in texture. But the absence of micas is a striking contrast, and hence its allocation to a different class is justified.

VI. Chemical Analyses.

(Analysed by P. K. Ghosh.)

1. Normal granite from Cheesewring quarry.
2. Normal granite from Fox Tor (top).
3. Normal granite (? albitized) from Notter Tor (lower quarry).
4. Godaver granite, underlying peat, from north flank of Hill Tor.
5. Godaver granite (fresh) from the Godaver region, just south of the foot-bridge.
6. Fine granite, 300 yards north of Dozmary farmhouse.
7. Killas inclusion (biotite-hornfels type) in a dike-modification of Godaver granite from Swit quarry, Craddock Moor.
8. Killas inclusion (biotite-hornfels type) in Normal granite, Notter Tor.

	1.	2.	3.	4.	5.	6.	7.	8.
SiO_2 ...	70·96	71·25	71·66	72·99	73·01	74·16	54·82	53·63
TiO_2 ...	·0·36	0·20	0·32	0·13	0·11	trace	1·60	1·97
Al_2O_3 ...	15·92	15·72	15·31	15·33	15·26	14·68	16·75	17·03
Fe_2O_3 ...	0·20	0·16	0·19	nil	0·29	0·26	1·06	1·10
FeO ...	0·84	0·86	0·84	1·09	0·73	0·65	9·53	9·91
MnO ...	0·30	0·21	0·28	0·06	0·07	0·07	0·34	0·33
MgO ...	0·49	0·32	0·25	0·28	0·06	0·18	1·60	1·77
CaO	1·11	1·42	1·21	0·64	0·68	1·01	1·54	1·75
BaO ...	trace	trace	n.d.	n.d.	0·02	nil	n.d.	n.d.
Na_2O ...	3·53	3·56	4·31	2·96	3·18	2·51	3·84	3·75
K_2O	5·60	5·51	4·26	5·59	5·47	5·15	6·54	6·55
F	0·08	0·05	0·06	n.d.	0·03	0·11	n.d.	n.d.
$H_2O\pm$ ...	0·80	0·71	0·83	1·18	0·63	0·53	1·73	1·84
B_2O_3 ...	0·28	0·29	0·25	n.d.	0·73	0·82	n.d.	n.d.
P_2O_5 ...	0·03	0·32	0·32	0·18	0·21	0·27	0·71	0·82
V_2O_5 ...	trace	n.d.	n.d.	n.d.	trace	n.d.	n.d.	n.d.
CO_2	n.d.	n.d.	n.d.	trace	n.d.	n.d.	n.d.	n.d.
Total ...	100·50	100·58	100·09	100·43	100·48	100·40	100·06	100·45
Sp. gr. ...	2·64	2·63	2·65	2·59	2·62	2·59	—	—

The analyses of the granites show the progressive enrichment in silica-content in the younger types. As shown by Brammall, the same feature is displayed by Dartmoor granites. Analysis no. 3 shows a marked departure from the usual ratio of $K_2O : Na_2O$. The higher Na_2O-content is probably due to causes somewhat akin to secondary albitization (see p. 293).

VII. Results of 'Heavy Residue' Determination.

(Accessory Minerals.)

	'Normal'.	'Godaver'.	'Fine'.	Elvan.
Percentage by weight of minerals heavier than bromoform (sp. gr. = 2·9)	8·5—5·5	5—4·4	3	1·1—2·4
Number of rocks crushed ...	(5)	(3)	(3)	(3)

These results are in agreement with facts established by Brammall for the Dartmoor granite, viz. the younger types contain a progressively lower percentage of accessories.

The accessories of the two main granite types are enumerated below:

In the Normal granite.—Magnetite, ilmenite, pyrrhotine, cassiterite, rutile, brookite, anatase, tourmaline, biotite, muscovite, andalusite

(purple and colourless), corundum (yellow, colourless, bluish), zircon, fluorite, beryl (very limited), apatite.

In the Godaver granite.—All the species named above, except beryl, with the additional species lithionite. Yellow corundum and colourless andalusite have not been observed.

Large amounts of wolframite have been found in river-sands. This mineral is probably derived from lodes and is not a primary constituent of the granite.

My very best thanks are due to Professor W. W. Watts and Dr. J. W. Evans for their never-failing help and advice throughout the period of my work. I also gratefully acknowledge my indebtedness to Mr. J. G. C. Leech, Dr. H. F. Harwood, Dr. A. Brammall, Dr. S. K. Chatterjee, Mr. M. Chatterjee, Mr. G. S. Sweeting, and Mr. E. J. Tallin.

Explanation of Plates VI and VII.

[Photomicrographs in ordinary light.]

Fig. 1. A porphyritic crystal of microcline in Normal granite. The core is formed of a Carlsbad-twin (M_1 and m_1) followed by a narrow zone of small crystals and narrow strips of plagioclase (P); then follows another zone of microcline (m_2 and M_2) in reversed orientation relative to the core; the third zone of microcline is just represented in the figure (right-hand, top) by m_3. The potash-felspar has a frilled margin (right of the figure, bottom), and is intergrown with plates of muscovite growing out into networks and spongy masses at the contact with the felspar. (×21.) [p. 294.]

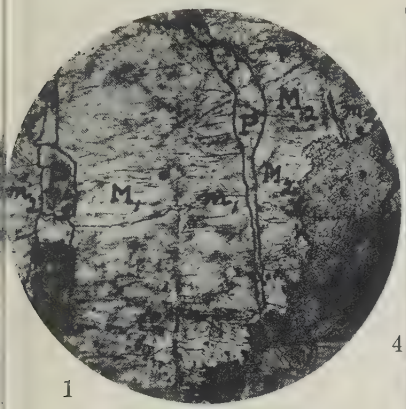
Fig. 2. Biotite-hornfels (banded type). It shows the alternate bands of biotite (dark) and granular plagioclase (light). (×26.) [p. 302.]

Fig. 3. Biotite-hornfels with a sub-ophitic texture. It shows biotite (b), set in a granular groundmass of quartz (Q), and plagioclase (P). (×17.) [p. 303.]

Fig. 4. The junction of granite with the microcline rim of a biotite-hornfels. The junction is towards the top where the biotite (b) appears. The biotite belongs to the granite and is arranged with its long axis parallel to the surface of contact with the rim (M); it contains inclusions of apatite (a). The rim contains inclusions of small crystals of plagioclase (P) with decomposed cores and hence appearing dark in the figure. Plagioclase decreases in amount farther from the contact. (×17.) [p. 303.]

Fig. 5. The same slide showing the junction of the microcline rim with the hornfels (the dark zone at the bottom). Note the abundant inclusions of quartz grains (Q) in the rim. (×17.) [p. 303.]

Fig. 6. The same slide (showing the hornfels only) farther removed from the granite contact. Note the advent of andalusite (A) as the view recedes from the contact. Compared with fig. 5, the texture is more open, and biotite is decidedly smaller in size as well as less idiomorphic. Andalusite is surrounded by a zone of muscovite (m) which is followed by plagioclase and biotite. (×17.) [p. 304.]



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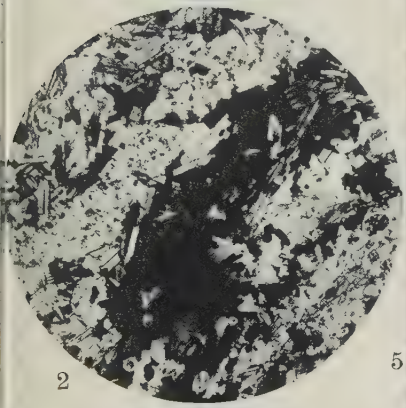

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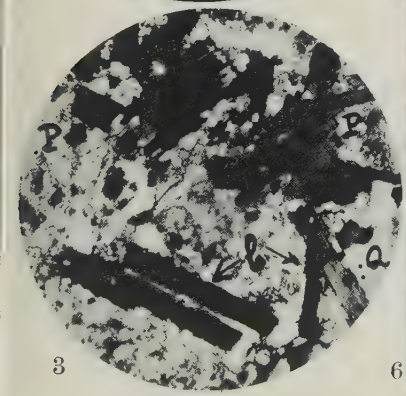

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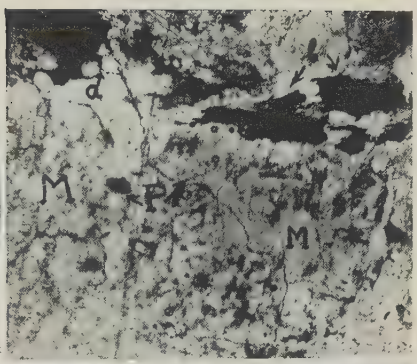

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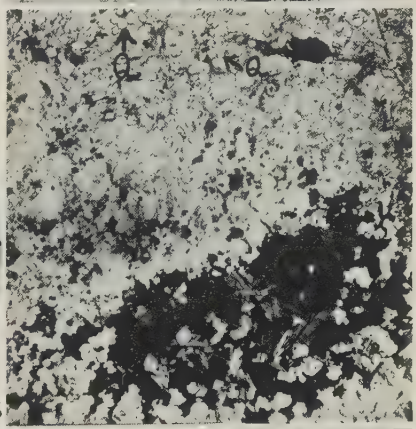

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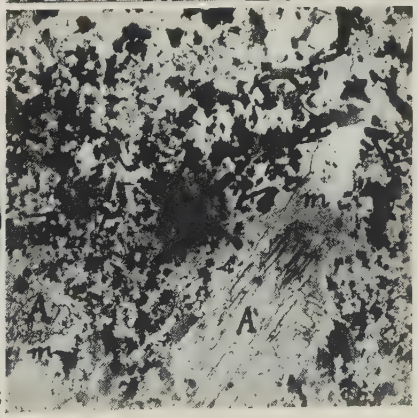

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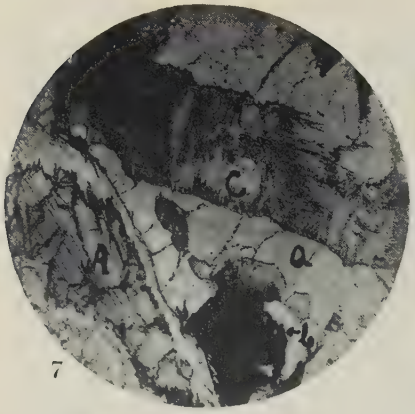
P. K. Ghosh: Bodmin Moor Granite and Hornfels Inclusions.



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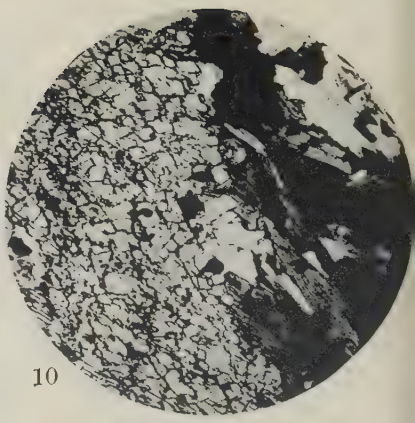

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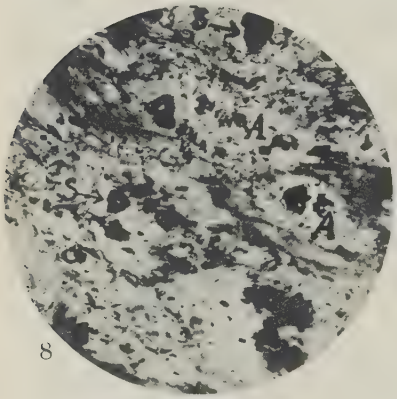

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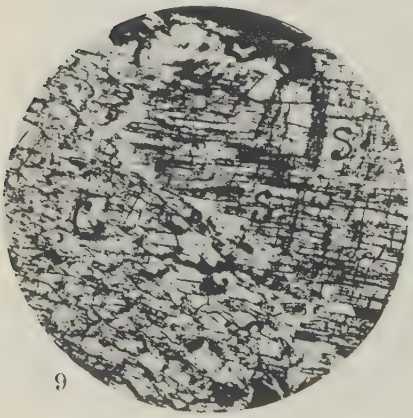

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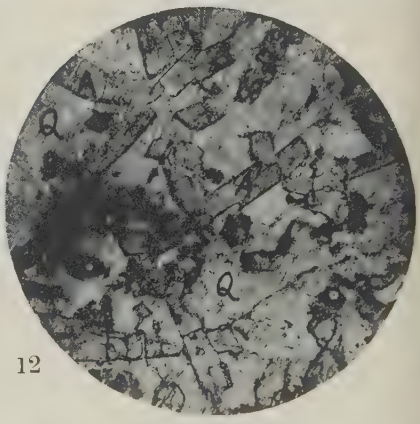

12

P. K. Ghosh: Hornfels Inclusions in Bodmin Moor Granite.

Fig. 7. Hornfels inclusion, showing porphyroblasts of pinitized cordierite (C) and andalusite (A), in a matrix of quartz (Q) and a little biotite (b). (×21.) [p. 305.]

Fig. 8. Hornfels inclusion, showing andalusite (A) with inclusions of spinel (S). Andalusite crystals are enmeshed in a tangle of biotite fibres. (×21.) [p. 305.]

Fig. 9. Hornfels inclusion showing porphyroblastic corundum (C) intergrown with sillimanite (S). The rest is biotite. (×21.) [p. 305.]

Fig. 10. The same showing large corundum (C) with well-developed pseudocleavage. There is also a band of biotite and quartz on the right of the figure. Small shreds of biotite occur along the partings of the corundum. (×21.) [p. 305.]

Fig. 11. Hornfels inclusion showing sheaf-like aggregates of sillimanite and shreds of biotite intergrown with one another; biotite appears dark in the figure. (×21.) [p. 305.]

Fig. 12. Hornfels inclusion showing plates of quartz (Q) ophitically enclosing andalusite (A) and plagioclase: the plagioclase is centrally decomposed and appears dark in the figure. (×21.) [p. 306.]

On the distribution of purple zircon in British sedimentary rocks.

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[Read June 14, 1927.]

I. *Introduction.*

BY the publication of his paper on 'The source of the purple zircons in the sedimentary rocks of Scotland',[1] Dr. Wm. Mackie has done his fellow-workers on the petrology of British sedimentary rocks a signal service. He has proved that this distinctive purple (or sometimes rose-coloured) variety of zircon is relatively abundant in the Lewisian gneiss. The variety occurs usually as rounded or ovoid grains, but occasionally with crystalline form; it is often finely polished, as if by wind, and is sometimes zoned, with a darker centre, or displays colour of varying intensity; it is often definitely, although but slightly, pleochroic, and usually possesses high birefringence. These characteristics at once bring to mind grains of zircon frequently met with in sedimentary rocks and recorded by numerous other observers.[2]

Dr. Mackie noted the presence of this variety as a detrital constituent in the Moine Schists, the Eastern Quartzites and Schists, the Torridonian Sandstone (and in pebbles of grit and quartzite in that rock), the Cambrian quartzites, limestones, and dolomites from north-western Scotland, the Old Red Sandstone of Scotland, the Carboniferous rocks in a boring near Glasgow, the Millstone Grit of Yorkshire, the Permian of Dumfries, and in the *Gordonia*-beds and the Triassic and Jurassic (?) sediments near Elgin. Moreover, he found the same purplish variety in various

[1] W. Mackie, Trans. Edinburgh Geol. Soc., 1923, vol. 11, pp. 200–213. [Min. Abstr., vol. 2, p. 311.]

[2] For example, Dr. R. H. Rastall refers to 'pink zonary zircons' in the Lower Greensand (Geol. Mag., 1919, vol. 56, pp. 218, 220, &c.). Other writers in their accounts of sedimentary rocks have referred certain purple zircon-like grains to xenotime. In my own notes from 1912 onwards, I have frequently recorded a 'pink or purple zircon-like mineral'. Since the publication of Dr. Mackie's paper, the references to the purple variety of zircon have been numerous.

specimens of pre-Cambrian gneisses and in a sedimentary rock from Canada, as well as in the Transvaal banket. But not the least interesting of his observations was that the true purple zircons are not common in the northern Scottish granites, other greenish, brown, and purplish-brown varieties being predominant. He suggested that the few purple grains found may, like andalusite, sillimanite, kyanite, staurolite, and garnet, have been picked up by the granite magma.

More recently, Dr. Mackie has deduced,[1] from the occurrence of purple zircons, that the Moine Series is more ancient than the Torridonian and that the latter rocks were derived in part from the Archaean, and in part from the Moine Schists.

That purple zircon is not typical of granites will, I think, be generally agreed. There is, however, a purplish-brown variety of more frequent occurrence in igneous rocks,[2] and which is often found in sediments. I am inclined to draw a distinction between the two varieties; the notes which follow, therefore, refer only to good rose-pink or purple grains.

The presence of this almandine-coloured variety of zircon is so widespread in sedimentary rocks and the type is so characteristic of certain British stratigraphical horizons, that the following notes, made in the course of other investigations during the past fifteen years, have been collected together in the hope that they may be of interest.

II. *The Coloration.*

Attempts have frequently been made to determine the probable source of different species and varieties of well-known rock-forming minerals by noting the special characters displayed by them, but it cannot be said that such attempts have always been successful. In this connexion the papers on zircon by G. Uzielli,[3] K. v. Chrustschoff,[4] H. Thürach,[5] G.

[1] Paper in course of publication in the Trans. Edinburgh Geol. Soc. (Abstract in Geol. Mag., 1927, vol. 64, pp. 141–142).

[2] For example, Dr. A. Brammall records it in the Dartmoor granite (Min. Mag., 1923, vol. 20, pp. 27–31). Subsequent investigations have led him also to record the presence of purple zircon, but the variety is not of common occurrence, and is confined to one phase of the granite.

[3] G. Uzielli, Sopra lo zircone della Costa Tirrena. Atti R. Accad. Lincei, Roma, 1876, ser. 2, vol. 3, pp. 862–877.

[4] K. v. Chrustschoff, Beitrag zur Kenntnis der Zirkone in Gesteinen. Tschermaks Min. Petr. Mitt., 1886, vol. 7, pp. 423–442.

[5] H. Thürach, Ueber das Vorkommen mikroskopischer Zirkone und Titan-Mineralien in den Gesteinen. Verh. Phys.-Med. Gesell. Würzburg, 1884, n. ser., vol. 18, pp. 203–284.

Spezia,[1] and J. Zerndt,[2] for example, may be cited. Curiously, perhaps, none of these observers except Zerndt seems to have referred to the rose-purple variety. Uzielli dealt with the probable source of some zircons and accompanying minerals, and gave detailed descriptions of their crystallography, but the only colour he noted was bluish. The work of Chrustschoff is too well-known to need more than passing reference. He mentioned grains which were colourless (' water-clear '), dusky, or of various tints of brown and yellow. Thürach, in his well-known paper cited above, gave an account of the crystal-forms and habits, but referred only to colourless, grey, and yellow varieties. Spezia attributed the red (presumably hyacinth tint) and brown coloration of zircon to the presence of iron and not organic substances, and expressed the view that colour-change does not depend on the temperature to which zircon is heated, and that thermoluminescence (' pyrophosphorescence ' as it was then called) is not related to the colour. These conclusions were contested by K. v. Kraatz-Koschlau and L. Wöhler,[3] who found that a loss of weight occurred in zircon heated in a stream of oxygen, and that carbon dioxide was found by passing the gases into lime-water. Spezia later replied (in the 1899 paper cited) to these criticisms, reaffirming his opinion that red zircon from Ceylon owed its colour to the state of oxidation of the iron present and not to organic matter, an insufficient quantity of which was present. He claimed that he had obtained an increase in the depth of colour on heating the mineral under oxidizing conditions, and a decrease on heating under reducing conditions.

A notable contribution to the literature of the colour of zircons is a more recent paper by Lord Rayleigh,[4] in which he described the effects of radio-active treatment of brown and reddish (hyacinth) varieties, the colour of which had been destroyed by heating to 300° C. Both the colour and property of thermoluminescence (which also disappears on heating) are restored by such treatment. He therefore attributed the colour to the effects of included radio-active substances.

Numerous later workers have figured types of zircon grains, and have

[1] G. Spezia, Sul colore del zircone. Atti Accad. Sci. Torino, 1876, vol. 12, pp. 37–43, and 1899, vol. 34, pp. 906–910.

[2] J. Zerndt, Über mikroskopische Zirkone aus den Karpathensandsteinen der Umgegend von Ciężkowice. Bull. Intern. Acad. Polonaise Sci. Lett., Ser. A, 1924, pp. 219–229.

[3] K. v. Kraatz-Koschlau and L. Wöhler, Die natürlichen Färbungen der Mineralien. Tschermaks Min. Petr. Mitt., 1899, vol. 18, pp. 304–333.

[4] R. J. Strutt, Note on the colour of zircons, and its radioactive origin. Proc. Roy. Soc. London, Ser. A, 1914, vol. 89, pp. 405–407.

attempted, like some of the investigators mentioned above, to refer them to sources in igneous or metamorphic rocks respectively. Others have endeavoured to show that such essays are of little value. Of late (perhaps wisely), these attempts appear to have been abandoned.

It seemed to me desirable to obtain, if possible, further information regarding the colour of the rose-purple variety, despite the difficulties involved in the small size and sporadic occurrence of the grains in sediments. To this end, special separations were made of the heavy detrital minerals from the Carboniferous Sandstone (Millstone Grit) from Glenboig, Lanarkshire, in which purple zircons are very abundant. In view of the possible confusion of brown or purple zircon with xenotime in sediments, spectroscopic examination was desirable, although Dr. Brammall had already proved that the brown zircons from the Dartmoor granite give no spectroscopic evidence of the presence of yttrium.

As I was unable to obtain with a spectroscopic attachment to the eye-piece of a microscope, a distinctive spectrum from even the most deeply-coloured purple grains, I am the more indebted to Messrs. R. W. Roberts and J. H. Hamshen of the Physics Department of the University of Liverpool for the trouble they undertook to obtain carbon-arc photographs of the absorption-spectrum of some typical purple grains. The results were negative in so far that no spectrum specially characteristic of this variety of zircon was obtained, but the failure may be attributed to the small size of the grains (0·1 to 0·2 mm. diameter). If larger crystals could be found, it would be more satisfactory to have the experiments repeated.

Dr. Mackie had already observed that the crystals of purple zircon were bleached, like other varieties of the mineral, on being heated in air to about 300° C. After treating some grains in this way (under oxidizing conditions), and rendering them colourless, I mounted a sample on a glass slip by means of an exceedingly thin smear of paraffin-wax, taking care that the grains were not covered by wax. Mr. R. W. Roberts then subjected these grains to the action of β- and γ-rays for 166 hours. The colour was partially restored, most of the grains showing a pale-pink or pale-purple tint. Presumably the duration of the treatment was insufficiently long for the restoration of the deep-purple colour.

III. *Distribution of purple zircons.*

The following notes relating to the occurrence of rose-pink or rich-purple zircon in British sediments are in no sense complete, but they may serve to stimulate investigation and lead to further recording of this

variety. It is desirable also that its associated minerals, which are often themselves distinctive, should be recorded.

As the number of individual localities is very large, they have been grouped in counties or districts.

Pre-Cambrian: Moine Schists; Eastern Schists (Scotland), Eastern Quartzites (Scotland); Torridon Sandstone, Quartzites (Donegal).

Cambrian: Shropshire; north-western Scotland.

Ordovician: ?

Silurian: Llandovery Sandstone of Shropshire and Malvern district. Denbigh Grit.

Old Red Sandstone: Northern Scotland; Kincardineshire, Perthshire; South Wales, Herefordshire, Shropshire.

Carboniferous: Throughout the whole system, including Lower Limestone Series, Calciferous Sandstone, Yoredales, Pendlesides, Millstone Grit, Lower, Middle, and Upper Coal Measures. Northern Ireland; Scottish Lowlands; I. of Arran; Northumberland, Durham, Yorkshire, Derbyshire, Midlands generally, Lancashire; North Wales (including Vale of Clwyd).

Permian: Elgin *Gordonia* Beds; Scottish Lowlands, Dumfries: Lancashire, Yorkshire, Midlands.

Bunter: North-eastern Ireland; North Wales (including Vale of Clwyd); Lancashire, Cheshire, Staffordshire, Warwickshire, Worcestershire, Gloucestershire, Nottinghamshire, Derbyshire, Yorkshire.

Keuper: Midlands, Cheshire.

Lias: Midlands, Cotteswold area, Dorset.

Inferior Oolite and Estuarine Series: Cotteswolds (northern end), Northampton Ironstone generally, Yorkshire.

Callovian: Yorkshire, Northamptonshire, Brora.

Corallian: Berkshire.

Kimmeridgian: Buckinghamshire, Oxfordshire, Berkshire, Dorset, Wiltshire, Lincolnshire.

Portlandian: Buckinghamshire, Oxfordshire, Dorset, Wiltshire.

Neocomian and Wealden: Spilsby Sandstone, Carstone of Yorkshire and Lincolnshire, Weald generally.

Aptian: Lower Greensand generally. Weald; Isle of Wight; Berkshire, Oxfordshire, Buckinghamshire, Bedfordshire, Norfolk.

Albian: Devon, Wiltshire; Weald; borings through London Basin. Also Belgium.

Chalk: East of England generally.

Eocene: All divisions from Reading Beds to Bagshot Beds, London Basin throughout; Isle of Wight and Hampshire; Dorset, Devon, and Cornwall. Also Belgium.

Pliocene: Diestian generally. Red Crag, Norwich Crag, Chillesford Beds, &c.; Cornwall. Also Belgium.

Pleistocene: Glacial and fluviatile deposits throughout England and Scotland.

Recent: Shore- and Dune-sands and River-deposits generally.

Although there are certain inequalities of distribution of purple zircons in the various formations listed above, these may be expected to disappear on the completion of further work. It may also be found that

formations not mentioned in the above table may prove to contain this variety of the mineral in addition to the other varieties known to be present. But there are certain British deposits in which purple zircon may be considered an abundant and characteristic constituent of the sediments and in which it is accompanied by other significant detrital minerals. Such deposits are the various divisions of the Carboniferous, Permian, and Bunter rocks, and the Lower Greensand.

In the Carboniferous rocks of Scotland and northern Ireland, purple zircons are found in great abundance, and are often of a typical deep colour and ovoid form. They are characteristic of the Carboniferous Limestone Series at Ballycastle, Caldwell, Campbeltown, I. of Arran, Monkredding, &c.; of the Upper Calciferous Sandstone Series at Coolkeeragh and Cookstown, of the Upper Limestone Series at Auchenheath; of the Millstone Grit at Blochairn, Crofthead, Drumcavil, Garngad, Garnkirk, Gartverrie, Germiston, Glenboig, &c.: and of the Coal Measures at Plean, Pordovan, Rawyards, &c.

The variety is often accompanied by abundant monazite, as at Auchenheath, Crofthead, Drumcavil, Garngad, Glenboig, Plean, Pordovan, Rawyards, or by staurolite, garnet, red rutile, chlorite, sphene, hornblende, pleonaste, and rarely kyanite. The proximity of the sediments to the ancient rocks of the Scottish Highlands leaves little doubt that all these minerals have a northern provenance. As the Millstone Grit and Coal Measures are traced southwards into Lancashire and Yorkshire, the stream of purple zircon and monazite can be followed with ease, but staurolite becomes less abundant and kyanite disappears.

Although there is a possibility of enrichment of the Midland Carboniferous rocks in monazite from Lake District or other granites, the trail of minerals of presumed Scottish provenance can be traced into Derbyshire, Warwickshire, and Staffordshire. For example, in the Millstone Grit of Biddulph Moor, monazite and purple zircon are plentiful.

In the Permian rocks of the north of England, purple zircons are by no means uncommon, their associates being monazite, staurolite, tourmaline, rutile, hypersthene, garnet, epidote, chlorite, and hornblende. Doubtless some of the Permian detrital material was obtained from older sediments, especially those of Carboniferous age, but it seems probable that some of it came direct from crystalline rocks. Thus the purple zircons of the Permian rocks of Lancashire and Yorkshire may be of 'composite' origin, but, since the grains are not so abundant as in the Carboniferous and are often of small size, these may in large part have passed through more than a single sedimentary cycle.

The purple variety of zircon may fairly be regarded as a characteristic and common constituent of Triassic sediments. It appears to be less abundant, however, in the Keuper than in the Bunter rocks. In the latter, the variety is found throughout Yorkshire, Derbyshire, and Nottinghamshire (Armthorpe, Burghwallis, Hempshill, Northallerton, Snaith, Whitley Bridge, Worksop, &c.), the Midlands (Warwickshire, Worcestershire, and Staffordshire), the Welsh borders, Lancashire, Cheshire, the Vale of Clwyd, Cumberland, Antrim, and Arran. It is often of a deep colour, of comparatively large size, and is associated with monazite, garnet, hypersthene, staurolite, sphene, hornblende, chlorite, deep-red rutile, pleonaste, epidote, and kyanite. These features suggest that it was directly derived from crystalline rocks, and not from Palaeozoic sediments.

Although purple zircon is occasionally seen in British Jurassic residues, it cannot be called an abundant or characteristic constituent. Its next re-appearance in quantity is in the Lower Greensand, where large ovoid deep-coloured and rose-pink grains are often abundant. At this horizon, also, it is accompanied by a flood of 'new' minerals, of relatively large size, the source of which cannot be placed in any British sedimentary rocks known to us. These minerals include abundant tourmaline (of varying colour and form), deep-red rutile, occasional garnets, pleonaste, sillimanite, andalusite, staurolite, sphene, monazite, chloritoid, epidote, and kyanite. Large and irregular grains of purplish-red rutile are especially noteworthy. The localities where purple zircon is plentiful are almost too numerous to mention, but they range throughout the outcrop north and south of the Weald, the Isle of Wight, Berkshire, Oxfordshire, Bedfordshire, Buckinghamshire, Cambridgeshire, and Norfolk. Again, the assemblage of minerals points to derivation from recently-exposed crystalline rocks.

Throughout the Upper Cretaceous and Tertiary strata, purple zircon is occasionally met with, but although in the Pliocene and Glacial deposits of East Anglia rich assemblages of minerals are found, the variety under consideration is neither an abundant nor typical constituent.

It is only to be expected that purple zircon should be of frequent occurrence in present-day shore- and river-sands. Large deep-coloured grains are abundant in 'concentrates' from such widely-separated localities as Montevideo, Tampico, Arkansas River, New Guinea, Senegambia, and Yunan (Korea). Many of these are from the neighbourhood of shields of Archaean rocks, which raises the question as to whether purple zircon

is a widespread constituent of the latter. It will be recalled that Dr. Mackie found the variety in the Transvaal banket and the 'Laurentian' gneiss of Canada.

Summarizing, therefore, we are able to say that the purple or rose-coloured variety of zircon is especially characteristic of those British sediments in which minerals derived from regionally-metamorphosed rocks are abundant, and of 'fresh' appearance. They provide additional evidence that the deposits were formed by denudation of land-masses composed of crystalline metamorphic rocks.

The separation of manganese in rock analysis.

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THE separation and estimation of small quantities of manganese in the course of exact analysis of igneous and metamorphic rocks has long presented a difficulty. The element is almost universally present, in amounts varying from a few hundredths of one per cent. to over one per cent., the higher figure being reached in the basic rocks, especially in those rich in ferromagnesian minerals. In the ordinary course of analysis, if no special method be adopted to separate and remove manganese, this element tends to be distributed between the 'ammonia precipitate' and the lime and magnesia. It has been shown by G. Steiger[1] that when the usual methods are employed, the manganese tends to be distributed in a quite haphazard way among the three precipitates; no definite proportion is retained by any one of them, although as a general rule most of the manganese is found with the magnesia and least with the lime. In those cases where the total amount of manganese present in the rock is small (0·1 % or less) no serious error is involved in disregarding its presence in the three precipitates, yet when larger amounts are in question, and especially in all cases where results of the highest accuracy are desirable, it is necessary to make some modification of the usual analytical procedure in order to permit of the concentration of the manganese in one particular precipitate, or, alternatively, to ascertain its distribution amongst the weighed precipitates of lime, magnesia, and alumina, so that a correction can be subsequently introduced. The present paper is an attempt to furnish a solution of this problem. Several methods have previously been suggested to achieve the above-mentioned result: they are outlined, and briefly criticized below:

1. Precipitation of the manganese by ammonium sulphide after removal of ferric oxide, alumina, &c., by means of ammonia.

[1] W. F. Hillebrand, The analysis of silicate and carbonate rocks. Bull. U.S. Geol. Survey, 1907, no. 305, p. 96.

2. Precipitation of the ferric oxide and alumina by the basic acetate method, followed by separation of the manganese as in (1) or by means of sodium acetate and bromine.

3. The manganese present in the lime and magnesia precipitates is determined colorimetrically in the solutions of the ignited and weighed precipitates. The total manganese present in the rock is determined colorimetrically in a separate weighed quantity, and the manganese which has been co-precipitated with the alumina is thus found by difference. The necessary corrections are then applied to all three precipitates.

4. Precipitation of the manganese quantitatively with the alumina precipitate, by the employment of an oxidising agent in the presence of ammonia.

The first method yields accurate results, but the removal of the manganese as sulphide is tedious, as the precipitate must stand overnight before filtration. Filtration itself is apt to be slow, and the excess of ammonium sulphide must be removed from the filtrate, so that the time required for carrying out an analysis is appreciably lengthened by these operations. Method (2) may be employed in the analysis of manganese ores, where the greatest accuracy is not required. It is not suited for use in rock analysis, as the precipitation of ferric oxide and alumina is never quite complete, especially when alumina predominates largely, as is usually the case with igneous rocks; also if the manganese in the filtrates be precipitated by means of sodium acetate and bromine, appreciable quantities of calcium are co-precipitated. The third method involves three separate determinations of manganese, but is fairly satisfactory, apart from the expenditure of time required. Its main objections are, that no determination of strontia in the lime precipitate can be made, and it is not possible to determine the amount of calcium invariably to be found in the magnesia precipitate. There is also some doubt as to the exact condition in which the manganese is present in the ignited magnesia precipitate: manganese ammonium phosphate is unstable in ammoniacal solution of the strength necessary for precipitating the magnesium, and consequently some manganese peroxide is always formed, a precipitate of magnesia which contains manganese being coloured brownish after ignition. The fourth method appears to be the most promising, as there is no difficulty in ensuring the complete precipitation of the whole of the manganese with the alumina by the simultaneous addition of an oxidizing agent: the objection to the method in its existing forms lies in the fact that small amounts of the other bases present, especially lime, tend to be carried

down in the precipitate in the form of manganites, and these are not wholly removed even by a second precipitation.

The present paper deals with a critical examination of the various oxidizing agents suitable, and their employment under varying conditions, with the object of seeing if it might not be possible to reduce the co-precipitation of lime and magnesia to a negligible minimum, or to obviate it entirely. The investigation has been confined to solutions containing from 1 to 50 milligrams of manganese, as a method which functioned satisfactorily between the above limits would prove applicable in practically all cases of rock analysis. Of the various oxidizing agents which suggested themselves as suitable, ammonium persulphate, hydrogen peroxide, chlorine, and bromine were tried. The use of the first of these has been previously proposed in the United States Geological Survey, Bulletin no. 700 (1919, p. 111), for this purpose, and has been adopted by H. S. Washington in his book on rock analysis.[1] The experience of the present authors in regard to this method was unfavourable. It was found impossible, even by a double precipitation, to obtain the whole of the calcium in the filtrate, and the presence of the relatively large amount of sulphate introduced into the analysis is also objectionable. The second reagent, hydrogen peroxide, would be particularly satisfactory if applicable, as it does not introduce any new ions into the solution, and any excess is easily removed by boiling. An extended series of experiments, in which the peroxide was added either prior to, with, or subsequent to the ammonia showed, however, that although the precipitation of the manganese with the alumina was quantitative, the precipitate invariably contained appreciable amounts of lime which could not be removed completely even after two precipitations. The use of chlorine as oxidizing agent gave similar results: the chlorine was introduced in two ways, either in the form of sodium hypochlorite, or by boiling the acidified solution with potassium chlorate, and then neutralizing with ammonia whilst Cl_2 and ClO_2 were being evolved. The precipitation of the manganese was complete under these conditions, but in every case considerable quantities of calcium were carried down in the precipitate.

Preliminary experiments with bromine as oxidizing agent gave more promising results, and it was finally decided to investigate this method quantitatively. For this purpose a number of carefully standardized solutions of the chlorides of aluminium, iron, calcium, and magnesium were prepared. Equal volumes of these solutions were then mixed, and

[1] H. S. Washington, Manual of the chemical analysis of rocks. 3rd edition, 1919, p. 151.

portions of 25 c.c. of the resulting solution used for each experiment. The strength of the various solutions was chosen so that the final liquid would contain amounts of the constituents similar to those which would be present in the solution obtained during the analysis of an average rock; a typical solution contained, in the form of chlorides, Al_2O_3 0·175, Fe_2O_3 0·10, CaO 0·075, MgO 0·0375 gram. The requisite amount of manganese was then added in the form of a standardized solution of manganese chloride, and the separation carried out. The precipitate of ferric oxide, alumina, and manganese oxide was filtered off, redissolved, and reprecipitated. In the united filtrates the lime and magnesia were determined by the usual methods.[1] Both these precipitates were redissolved after being weighed, and the resulting solutions tested for manganese by the periodate method, but in no case was this element found in more than unweighable traces, and usually it was entirely absent. In all cases where the separation proved satisfactory, the experiments were repeated after varying the amounts of the other constituents, in order to eliminate possible errors due to special conditions.

Details of the method as finally adopted, are given below. The solution should have a volume of about 400 c.c., and sufficient HCl must be present to prevent the precipitation of magnesia on the addition of ammonia. The liquid is heated to boiling, and ammonia added in the usual way in slight excess. Bromine water is then run in by means of a tap funnel, the stem of which is drawn out to a fine jet, and small additional quantities of ammonia are simultaneously added, in order to keep the solution alkaline.[2] When all the bromine has been added, a few more drops of ammonia are added, the solution boiled, and the precipitate filtered off. It is redissolved in dilute HCl containing a little H_2O_2, and the precipitation repeated exactly as before. The quantity of bromine required depends on the amount of manganese present; suitable amounts are shown below:

10	mgms.	of	MnO	require	the	addition	of 15 c.c. saturated Br water.
50	,,	,,	,,	,,	40	,,	,,
100	,,	,,	,,	,,	75	,,	,,

The results of two typical series of experiments carried out to determine the validity of the above method of precipitation are given in the

[1] After concentration of the solution and removal of the small amount of alumina still present (vide W. F. Hillebrand, The analysis of silicate and carbonate rocks. Bull. U.S. Geol. Survey, 1919, no. 700, p. 114).

[2] Free acid is produced in the reaction, and it is essential to add sufficient ammonia to ensure the liquid remaining alkaline throughout the precipitation.

annexed tables. In all cases a double precipitation with ammonia and bromine was made. The solution used contained, in addition to the quantities of lime and magnesia shown in the tables, Al_2O_3 0·175 and Fe_2O_3 0·1 gram as the chlorides of these metals. It will be seen that the separation of the manganese is quite satisfactory up to a limit of about 50 milligrams MnO; above this they are liable to be irregular, some co-precipitation of lime and magnesia resulting. The above limit is, however, ample as regards the quantities of manganese met with in the analysis of igneous rocks, which is the particular case that the method is devised to cover.

In order to ascertain that the whole of the manganese introduced into the solution was recoverable from the ammonia precipitates, those obtained in the second series of experiments were dissolved in HCl, taken down to strong fuming with sulphuric acid, and the manganese in the resulting solution determined by the bismuthate method. The following results were obtained:

MnO originally present in solution.	MnO found in the ammonia precipitate.
0·0049 gram	0·0044 gram
0·0098	0·0095
0·0197	0·0197
0·0492	0·0488

Series I.

No. of Experiment.	Amount MnO (added).	Amount CaO (found).	Amount $Mg_2P_2O_7$ (found).
Check A	none	0·0637 gr.	0·1425 gr.
Check B	none	0·0634	0·1437
Check C	none	—	0·1425
1.	0·0010 gr.	0·0639	0·1420
2.	0·0025	0·0632	0·1438
3.	0·0053	0·0635	0·1421
4.	0·0053	0·0630	0·1428
5.	0·0101	0·0639	0·1432
6.	0·0101	0·0638	det. spoilt
7.	0·0201	0·0632	0·1428
8.	0·0201	0·0634	0·1422
9.	0·0503	0·0635	0·1414
10.	0·0503	0·0638	0·1424

Series II.

No. of Experiment.	Amount MnO (added).	Amount CaO (found).	Amount $Mg_2P_2O_7$ (found).
Check A	none	0·2102 gr.	0·1754 gr.
Check B	none	0·2108	0·1757
1.	0·0049 gr.	0·2099	0·1768
2.	0·0098	0·2108	0·1754
3.	0·0197	0·2113	0·1759
4.	0·0492	0·2105	0·1751
5.	0·0492	0·2102	0·1753
6.	0·0738	0·2012	0·1767
7.	0·0738	0·2104	0·1539
8.	0·0738	0·2100	0·1669
9.	0·0984	0·2010	0·1762
10.	0·0984	0·2086	0·1722
11.	0·0984	0·2071	0·1705
12.	0·0984	0·2079	0·1746
13.	0·0984	0·2096	—
14.	0·0984	0·2097	—

A new model refractometer for determining the refractive indices of gem-stones, crystals, and liquids.

By Bristow J. Tully, F.G.S., F.Z.S.

[Read November 3, 1925.]

THE general outline and weight of the body of the instrument are devised to ensure perfect rigidity and steadiness during use, all necessary adjustments, such as the focusing of the rotating eyepiece, movement of the revolving stage, reflector, or screen, being quite independent of the optical system; which, being contained in the main body in a dust-proof receptacle, entirely prevents any vital part of the refractometer getting out of adjustment, unless deliberately tampered with.

The eyepiece, which by rotation is capable of giving a wide variation of focus, is set at the most convenient angle to ensure easy and comfortable observation.

The forward projecting portion attached to the main body is rectangular in shape, and hollow, the upper surface forming a platform in which revolves a circular plated stage with a milled edge capable of being turned about a vertical axis through 360°, with an engraved scale marked at every 45° to enable the observer to read with accuracy the least and greatest refractive indices of all doubly refracting stones.

Into this circular stage is mounted the soft dense glass hemisphere *B*, and this stage being capable of rotation allows observation of a gem or crystal being made from all directions, thus obviating the frequent necessity of turning the specimen on the dense hemisphere, and consequent risk of scratching by the far harder specimens.

The velvet-lined shield *D*, hinged at the forward end of the platform, serves the dual purpose of acting as a screen to keep out all extraneous light that may enter from above during examination of a gem; and also as a protection to the hemisphere when the instrument is not in use.

The matt opal reflector *A*, hinged below the shield, affords a most suitable means of throwing a beam of diffused light to the under surface

of the hemisphere, thereby giving an even illumination to the scale. It is provided with a catch to hold it at about 45° when in use, or vertically when in the case. When it is desired to use monochromatic light, the reflector can be swung downwards out of range.

From the first, I intended the instrument to be essentially a 'stand' instrument, practical, comfortable in use, and stable upon a bench or

Fig. 1. The 'Tully' Refractometer.

table; means were to be provided for obtaining the refractive indices of doubly refracting substances without having to turn them, as in existing types, upon the comparatively soft glass hemisphere; the scale was to be well magnified and read upwards commensurate with increasing values, with the low numbers commencing at the bottom; and the optical performance had to be clear and definite.

After designing various models, in the final type the fulfilment of a number of these conditions went very conveniently hand in hand. To erect the eyepiece scale, it was necessary to make several reversals of the

beam of light, by lenses, mirrors, or by both, and a consequent lengthening of the optical system was inevitable. At the same time, comfortable positions for the head in observing, and for the hand in rotating the hemisphere were required, so that the eyepiece had to be some inches above the base of the instrument, and the hemisphere some inches in front of the eyepiece. This has been done by making the body of the instrument of two antimony-bronze castings—a main body and a projecting portion. The projecting portion carries the hemisphere an inch or so before the front face of the main body, and the eyepiece is adjusted at the back of its top face. The optical system entirely occupies the main body. The accuracy of the surfacing, placing, and form of the lenses give the required degree of optical performance.

Considerable difficulty was experienced in the satisfactory mounting of the hemisphere on a narrow ring of a flat plate so that the rotation took place on an axis through the centre of the hemisphere. This difficulty was overcome by fitting a very shallow ring to the plate, bedding the edge of the hemisphere and the ring in a special plaster, and centring up on a spindle during setting. The plate and hemisphere turn in machined grooves in the small front casting which is rigidly attached to the main body.

Only one external adjustment is necessary, namely, that of focusing the eyepiece upon the scale. The eyepiece and hemisphere are fixed in position with respect to the main body, and the adjustments for position and range of the shadow-edge are made by altering the positions of elements in the optical system during the assembly of the instrument. Three lenses and two prisms are employed. The first lens *F*, very close to the hemisphere, is contained in a sliding framework ; immediately behind, mounted in trunions, is the first prism *G*. Movement of the first lens along the optical axis with respect to the hemisphere makes the range of the shadow larger or smaller, e. g. with the 1·4 reading correct, the 1·7 reading might actually be 1·6 or 1·8. Movement of the first lens and prism provides a coarse adjustment for the position of the shadow-edge, e. g. the 1·4 reading might be at 1·3 on the scale, and the reading 1·72 at 1·62 ; the actual linear difference between 1·4 and 1·3 being about the same as that between 1·72 and 1·62. The first prism is mounted so that a small rotation about the mid-line of its hypotenuse can be made, by means of a screw, and this provides a fine adjustment for the position of the shadow-edge.

The second prism *I* comes between the second and third lenses, *H* and *J*, and reflects the light back along a parallel path. It plays no part in

the adjustments, so is fixed in position, and merely makes the optical system compact. The second and third lenses are both adjustable along the optical axis, and, in conjunction with the first lens, bring the shadow-edge clearly to the eyepiece scale. One lens actually would be sufficient to do this, but it would have to be placed opposite the middle of the

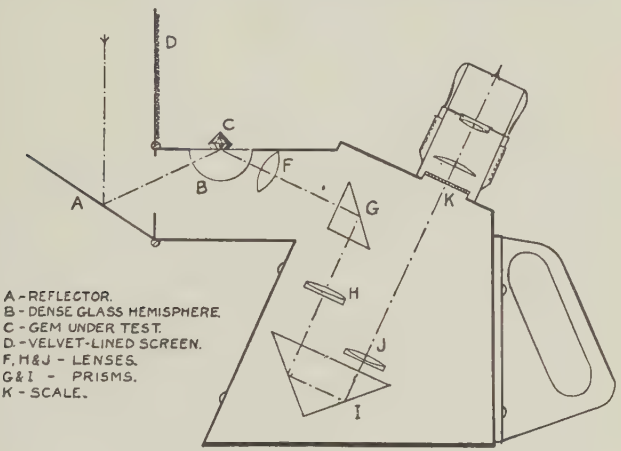

Fig. 2. Diagrammatic section through Refractometer.

second prism, and to be provided with means of adjustment, conditions rather difficult to fulfil.

A specially plotted photographic scale *K* showing the divisions very clearly is fixed under the compound rotating eyepiece.

The instrument is chiefly intended for the determination of the refractive index of gem-stones, and difficulties have arisen owing to the width of the dispersion band that is always met, say, in the case of spinel and sapphire. It becomes necessary either to set the instrument for a certain wave-length and leave the location of it in the spectrum seen to the user, or to cut off some part of the spectrum by a filter. If a filter could be used that allowed only the extreme red to pass through it, to all intents and purposes for every stone there would be a sharp black line, for not much red is seen. But so very little light would be enabled to pass through at all that under any but the best lighting conditions it would be very difficult to say which was the shadow part of the field and which was the light part. A compromise has been effected that seems to meet with approval. A light-orange

filter has been fitted over the eyepiece, and the shadow is seen with a green edge. The green edge is not very far from the sodium-yellow position, so that readings taken at the green edge will be found to agree almost exactly with those taken with monochromatic sodium-light, especially if a liquid with little dispersion, such as monobromonaphthalene, is used. If the available light is bad and the orange filter is removed, the first 'dark' part of the dispersion band is the green edge, so that agreement is again obtained.

It must be emphasized that this instrument is not intended for field work or for work of the high order of accuracy of the Abbe instrument. Refractive indices are marked on the scale to the second decimal place and can be read off with ease by estimation to the third place. A gem-stone or crystal on the hemisphere can be turned with comfort while examining the shadow-edge through the eyepiece ; and with the solution of the dispersion trouble, a definite addition in the field of mineralogical instruments would seem to have been supplied.

In conclusion, I wish to tender my sincere thanks to Mr. J. Pike, A.R.C.S., and Mr. Hobson, of the firm of Messrs. Rayner of 9 Vere Street, London, W. 1, the makers and co-patentees, for their help and technical suggestions in overcoming several difficulties arising during the evolution and subsequent completion of the model.

Corundum twins from Transvaal.

By L. J. Spencer, M.A., Sc.D., F.R.S.
Deputy-Keeper in the Mineral Department, British Museum (Natural History).

[Read June 14, 1927.]

LAMELLAR twinning is of frequent occurrence in corundum, as evidenced by rulings on the surfaces and by the well-marked pseudo-cleavages (or partings) parallel to the faces of the primary rhombohedron and also parallel to the basal plane. Distinctly developed juxtaposition and interpenetration twins would, however, appear to be rare, for only a few isolated examples have hitherto been described.

There are two twin-laws for corundum crystals, the twin-plane being either $r\,(10\bar{1}1) = (100)$ or $c\,(0001) = (111)$. The former of these is the one considered in the present note. Of these r-twins only six small ($\frac{1}{4}$ to 1 inch across) isolated crystals have previously been figured and described in detail. References to these are given under Barvíř, Bowman, and Nies and Goldschmidt in the following:

Bibliography of corundum twins (apart from lamellar twinning).[1]

P. V. Eremyeev, Verh. Russ. Min. Gesell., 1878, ser. 2, vol. 13, p. 440; 1879, vol. 14, p. 227; Gornyi Journ., 1887, vol. 3, p. 263. [Russian; Abstracts in Zeits. Kryst. Min., vol. 2, p. 505; vol. 3, p. 438; vol. 15, p. 537.] r- and c-twins, from Ural.

A. von Lasaulx, Zeits Kryst. Min., 1885, vol. 10, p. 362, pl. 12, figs. 12 and 13. r-twin, from Buncombe Co., North Carolina.

W. E. Hidden and H. S. Washington, Amer. Journ. Sci., 1887, ser. 3, vol. 33, p. 507. r-twin, from Ceylon.

H. Barvíř, Zwei Sapphirzwillinge. Ann. Naturhist. Hofmus. Wien, 1892, vol. 7, pp. 135–139, 3 figs. Two r-twins, from Ceylon.

M. Bauer, Neues Jahrb. Min., 1896, vol. ii, p. 213, fig. A. c-twin, from Burma.

O. Mügge, Tschermaks Min. Petr. Mitt., 1899, vol. 19, p. 165, 1 fig. c-twin of artificial corundum.

H. L. Bowman, A twin crystal of sapphire. Min. Mag., 1900, vol. 12, pp. 355–358, 1 fig. r-twin, from Kashmir?

[1] On lamellar twinning see, for example, J. W. Judd, On the structure-planes of corundum. Min. Mag., 1895, vol. 11, pp. 49–55.

G. Melczer, Math. Természettud. Értesítő, Budapest, 1901, vol. 19, p. 482, pl. 9, fig. 4, pl. 10, figs. 22–24 ; Zeits. Kryst. Min., 1902, vol. 35, p. 571, pl. 14, fig. 4, pl. 15, figs. 22–24. *c*-twin, from Burma ; *c*-twinning in artificial corundum shown by the reversed orientation of etch-figures on the basal plane.

W. E. Hidden, Corundum twins. Amer. Journ. Sci., 1902, ser. 4, vol. 13, p. 474, 1 fig. *c*-twins, from Macon Co., North Carolina.

R. Brauns, Neues Jahrb. Min., 1906, vol. i, p. 47, fig. 10. Twin on $d(10\bar{1}2)$?, from Ceylon.

A. Nies and V. Goldschmidt, Neues Jahrb. Min., 1908, vol. ii, pp. 108–112, pl. 9, fig. 3, pl. 10, figs. 4–5. Three *r*-twins, from Ceylon and Kashmir. A coloured picture of one of these small crystals in the Seligmann collection is given by R. Brauns in 'Das Mineralreich' 1903–4 ('The Mineral Kingdom', translated by L. J. Spencer, 1908–12), pl. 42, fig. 7.

A. L. Hall, Corundum in the northern and eastern Transvaal. Mem. Geol. Survey South Africa, Pretoria, 1920, no. 15, 223 pp. Mention of *r*-twins on pp. 66–68, 150, 155, pl. 17, fig. 8. [Min. Abstr., vol. 1, p. 210.]

With one exception, all the line-drawings given in the papers quoted above are reproduced in Professor V. Goldschmidt's monumental 'Atlas der Krystallformen'.[1] Of the 139 figures of corundum crystals there collected together, 16 represent twinned crystals, and of these 9 are of *r*-twins.

The twin-crystals of corundum to be here described have already been mentioned by Dr. A. L. Hall in his exhaustive memoir on corundum occurrences in the Transvaal. There he gave an accurate account of the essential features together with a photographic reproduction (though unfortunately rather poor and inadequate) of one of the twins. These details, of special interest to crystallographers and mineralogists, are, however, buried in a book of some size written from a geological and economic point of view, and they have been overlooked in notices and abstracts. Dr. Hall concurs with me that this matter merits more detailed notice, and for this purpose he has generously supplied me with abundant material for description.

The main material on which I started work was, however, independently supplied by a fortunate find that I made at the British Empire Exhibition at Wembley in 1925. There amongst the large economic display of corundum from the Transvaal I noticed a block of rock, weighing $33\frac{1}{2}$ lb., which showed the impression of a large twinned crystal, together with numerous other twins throughout the mass. Through the kind offices of Captain W. W. Luffman, who had been left in charge of the exhibits at the close of the Exhibition, this remarkable specimen was secured for the mineral collection of the British Museum.

[1] V. Goldschmidt, Atlas der Krystallformen, 1918, vol. 5, pls. 18–26. *r*-twins are shown in figs. 47, 56–59, 89, 119–121 ; *c*-twins in figs. 71, 88, 94, 96, 104–106.

Thinking that a corundum twin of this size (6 × 6 inches)[1] might be unique, a plaster mould and cast were first prepared and skilfully coloured by Mr. P. Stammwitz, preparator in the Department of Zoology. A photograph taken by Mr. H. G. Herring of this coloured model is re-

Fig. 1. Corundum twin from Transvaal. (Photograph of plaster cast.) Reduced to ½ natural size.

produced in fig. 1. The block of rock was then broken up, still preserving the large impression, with the object of exposing other twin-crystals.

The locality of the specimens, as described in Dr. Hall's memoir, is a corundum pit, the Turkaspost mine, on the Turkaspost farm (no. 941, formerly no. 1621), immediately to the west of Bandolier Kop railway station in Zoutpansberg district, northern Transvaal. The corundum-bearing rock represents modified apophyses of granite intrusive into

[1] Dr. A. L. Hall in his memoir (loc. cit., p. 67) mentions such twins measuring 6 inches across with a thickness of one-eighth of an inch, and he has informed me that twins of this size are by no means uncommon at the locality.

serpentine, pyroxenite, and other ultra-basic rocks. The corundum has the form of hexagonal prisms or irregular nodular masses, but in certain parts of the pit the platy form only is present.

This is the only locality mentioned by Dr. Hall for corundum of this type, but I am informed by Professor Charles Palache of Harvard University that while on a visit to South Africa in 1922 he collected specimens, practically identical with those from Turkaspost, from a small opening in the south-west corner of Waerkum farm (no. 717), about 10 miles SW. of Mara railway station, and about 20 miles NW. from Bandolier Kop.

The large (33½ lb.) block showed numerous large platy crystals of corundum penetrating the rock in all directions. The crystals are often in contact with one another, and they would appear to form a very open network with the snow-white coarsely-crystallized felspar filling the interspaces. This felspar was determined by Dr. Hall to be an acid plagioclase near andesine, and the rock he described as plumasite-pegmatite.[1] In this and in other similar specimens sent by Dr. Hall from Turkaspost no biotite was seen. Biotite is, however, often present in the plumasite from other localities in the Transvaal, and is, for example, well shown in a large block weighing 88½ lb. from Palmietfontein farm (no. 374) (10 miles north of Bandolier Kop) which was presented to the British Museum collection by Messrs. A. W. Collet and S. Snertinge in 1919; here the large corundum crystals are of the normal hexagonal habit. The rock usually breaks along the corundum crystals, leaving impressions of their striated and corrugated surfaces on the felspar; and on these felspar surfaces there are often black dendritic markings of manganese oxide or brown stains of limonite.

The corundum crystals are dark greyish-brown in colour and opaque in mass. Their faces are rough and corrugated, and unfortunately not suitable for measurement on the reflecting goniometer. All that could be obtained were approximate measurements with a protractor or a contact-goniometer of the angles between striae and the somewhat rounded edges. The crystals range from 1 to 5 mm. in thickness, being usually 2–3 mm.; and on the flat face they vary considerably in size, up to 6 inches (15 cm.) across for the large impression (fig. 1). As described by Dr. Hall these arrow-head twins have a face of the primary

[1] Plumasite was first described from Plumas Co., California, by A. C. Lawson in 1903 as an oligoclase-corundum rock. It has been formed by the desilicification of granitic magma intrusive into ultra-basic rocks. [Min. Abstr., vol. 1, pp. 16, 210, 282; vol. 2, p. 80; vol. 3, p. 84.]

rhombohedron, $r(10\bar{1}1) = (100)$, as twin-plane and are flattened parallel to the pair of prism-faces that are perpendicular to the twin-plane. This habit being so unusual for corundum and at first rather puzzling, an attempt has been made to represent it diagrammatically in figs. 2–5. These figures are simple elevations and plans, with (in the elevations) the twin-axis and the flattening of the crystals in the plane of the

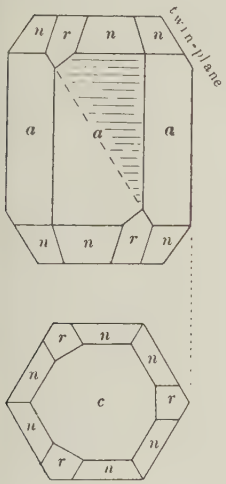

Fig. 2.

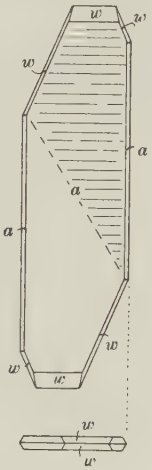

Fig. 3.

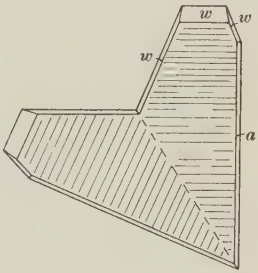

Fig. 4.

Twinning of corundum on $r(10\bar{1}1)$.

Elevations and plans: the twin-axis and flattening of the crystals in the plane of the paper.

paper, according to the method of drawing twinned rhombohedral crystals advocated by the late Professor W. J. Lewis.[1]

Fig. 2 represents the familiar text-book figure of a corundum crystal with the second-order hexagonal prism $a(11\bar{2}0) = (10\bar{1})$, basal pinakoid $c(0001) = (111)$, the primary rhombohedron $r(10\bar{1}1) = (100)$, and a hexagonal bipyramid $n(22\bar{4}3) = (31\bar{1})$. All these forms, with the exception of the base, were observed on the present crystals, but r is only slightly developed, and the hexagonal bipyramid is more often the steeper form $w(11\bar{2}1) = (14\bar{2})$. A crystal with only the planes a and w and flattened parallel to one pair of prism-faces is represented in fig. 3, from which, it is readily seen, the arrow-head twins (fig. 4) are derived by symmetrical repetition (reflection) across the dotted line which

[1] W. J. Lewis, Min. Mag., 1908, vol. 15, p. 62.

represents the trace of the twin-plane. The angle $cr = 57° 34'$ is the angle between the horizontal striations on a and the trace of the twin-plane, so that the angle between these striations in the two portions of the twin is $115° 8'$. Angles of 114–116° were measured between the two sets of striations. The outside edges of the arrow are bounded mainly by the prism-edges, and these also form an angle of $64° 52'$ ($180° - 115° 8'$). Usually, however, the point of the arrow presents a more acute angle; measurements gave values ranging from about 30° to 57° (the latter in fig. 1). The reason for this is an oscillation between a and r at the edges, as represented in fig. 5. The re-entrant angle at the opposite end of the arrow-head shows in fig. 1 a concave curvature and is evidently formed by a series of edges. In the few other complete examples the edges of this re-entrant angle are straighter and are bounded by narrow faces of the form $w(11\bar{2}1)$. Rough measurements of the angles between these edges and the twin-suture suggest that the following planes in adjacent pairs may form the bounding edges.

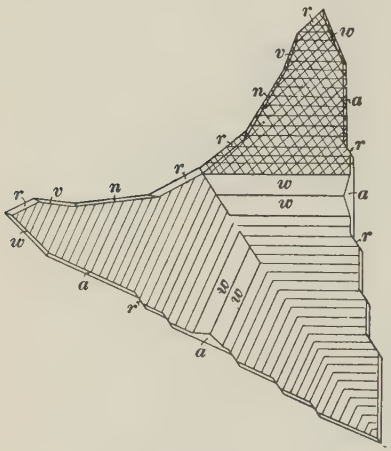

Fig. 5. Diagram of corundum twinned on $r(10\bar{1}1)$.

	Slope of edge between adjacent pair of planes.	Angle of edge to twin-suture: Calc.	Obs.
$-(2.2.\bar{4}.15) = (753)$	... 17° 28′ ...	75° 2′	76°
$r\ (10\bar{1}1) = (100)$ ...	... 38 12 ...	95 46	97–99
$n\ (22\bar{4}3) = (31\bar{1})$ ...	... 57 34 ...	115 8	117
$w\ (11\bar{2}1) = (41\bar{2})$ ...	... 67 2 ...	124 36	124–126
$v\ (44\bar{8}3) = (51\bar{3})$ ...	... 72 23 ...	129 57	129
$\theta\ (8.8.\bar{1}\bar{6}.3) = (91\bar{7})$	... 80 58 ...	138 32	137

As indicated in the figures the prism-faces are prominently striated horizontally, i. e. perpendicular to the prism-edges. This is due to an oscillation with the hexagonal bipyramid, and it sometimes takes the form of coarse corrugations. Angles of 42° and 30° measured over the ridges correspond with the pyramids $w(11\bar{2}1)$ and $v(44\bar{8}3)$ respectively.

In addition to these striations there are also extremely fine rulings in three directions on the prism-faces (represented in fig. 5 at the top right-hand corner). Those parallel to the coarse striations are due to lamellar twinning on *c*; another set parallel to the twin-suture is due to lamellar twinning on the *r*-plane which is the twin-plane; while the third set inclined at 95° 46′ to the twin-suture is due to lamellar twinning on one or other, or both, of the other two *r*-planes. This lamellar twinning gives rise to excellent planes of parting in the crystal, with smooth bright surfaces parallel to both *c* and *r*. As an aid in orientating the crystals it was found that flakes from the *c*-parting gave a central uniaxial optic figure, while flakes from the *r*-partings showed an optic axis at the edge of the field of view in the microscope. Owing to these partings and also to the thinness of the crystals they are very fragile; and for this reason, although some hundreds of crystals were present in the large block, only few more or less completely formed twins were successfully extracted. These measure from $2\frac{1}{2} \times 3\frac{1}{2}$ to 8×10 cm., and remain attached on one side to the rock; without this support the crystals break up into fragments. The fine rulings of the lamellar twinning are impressed on the felspar surfaces. The 'secondary' lamellar twinning was therefore present previous to the crystallization of the felspar; and were not, in this case, the result of later pressures in the rock. The crystals are occasionally bent or broken across, and this evidently happened before the loose aggregate of crystals received the support of the crystallized felspar. The residual melt (with the composition of oligoclase-andesine) left after the crystallization of the corundum was evidently very viscous.

The extreme flattening of these twin-crystals of corundum in a direction perpendicular to the twin-plane is very remarkable. As Dr. Hall has pointed out, the crystals may be as much as thirty to fifty times as wide as they are thick. Untwinned crystals of corundum from other parts of the same mine are equally developed in all directions. A similar but less pronounced flattening perpendicular to the twin-plane is also sometimes, though not always, shown by some other minerals, e. g. the Japanese-twins of quartz, butterfly-twins of calcite, and spinel-twins of diamond. No explanation of this flattening appears to have been given. The corundum twins previously described from Ceylon and Kashmir also show a tendency to the same flattening, though not to such a marked degree. The same is the case with most of the British Museum specimens mentioned below from these localities, but this is not invariably so, for some of the twinned crystals are equably developed.

Distortion in twin-crystals is discussed by H. Tertsch[1] in his recent volume on the habit of crystals.

Twins of corundum are evidently not so rare as previously supposed. At the Turkaspost, Transvaal, locality they could be collected by the thousand, or even by the ton. Also if searched for in collections they are to be found. In the British Museum mineral collection a small set of six well-developed crystals from Ceylon and Kashmir are specially displayed as examples of *r*-twins, and there is also an interesting specimen from Kashmir showing an intergrowth of an *r*-twin of corundum with a crystal of tourmaline. Further, by searching through the large series of isolated crystals from Ceylon several examples can be found showing evidences of twinning. Small crystals with much the appearance of buds can be seen branching out at 65° from the sides of the main crystal. My attention to this fact was first called by Mr. B. W. St. Clair of the General Electric Company at Lynn, Massachusetts, who handles considerable quantities of precious and semi-precious corundum for the purpose of making pivot-supports for electrical instruments. Recently, while in London, he secured a parcel of 1,000 ounces of Ceylon crystals and pebbles that had been rejected by the gem-cutters, but eminently suitable for grinding as pivot-supports. This lot I had the privilege of sorting over, and I was able to pick out eleven crystals showing indications of twinning. These eleven crystals weighed $\frac{2}{3}$ oz., representing 0·06 % of the whole parcel.

[1] H. Tertsch, Trachten der Kristalle. Berlin, 1926 [Min. Abstr., vol. 3, p. 223]. See also F. Becke, Über die Ausbildung der Zwillingskristalle. Fortschr. Min. Krist. Petr., 1911, vol. 1, pp. 68–85 ; and H. Hilton, The energy of twin-crystals. Min. Mag., 1909, vol. 15, pp. 245–246.

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There are four meetings of the Society each year, on Tuesdays at 5.30 p.m. in January, March, June, and November. They are held (by kind permission) in the Apartments of the Geological Society, Burlington House, Piccadilly, London, W. 1. Titles of papers to be read at these meetings, with a view to subsequent publication in the Magazine, should be sent to the General Secretary at least two weeks before the meeting. Authors alone are responsible for the views set forth in their respective papers. They are allowed, free of charge, fifty separate copies of their papers published in the Magazine.

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